

Research progress on the working mechanisms of QLEDs based on novel characterization methods

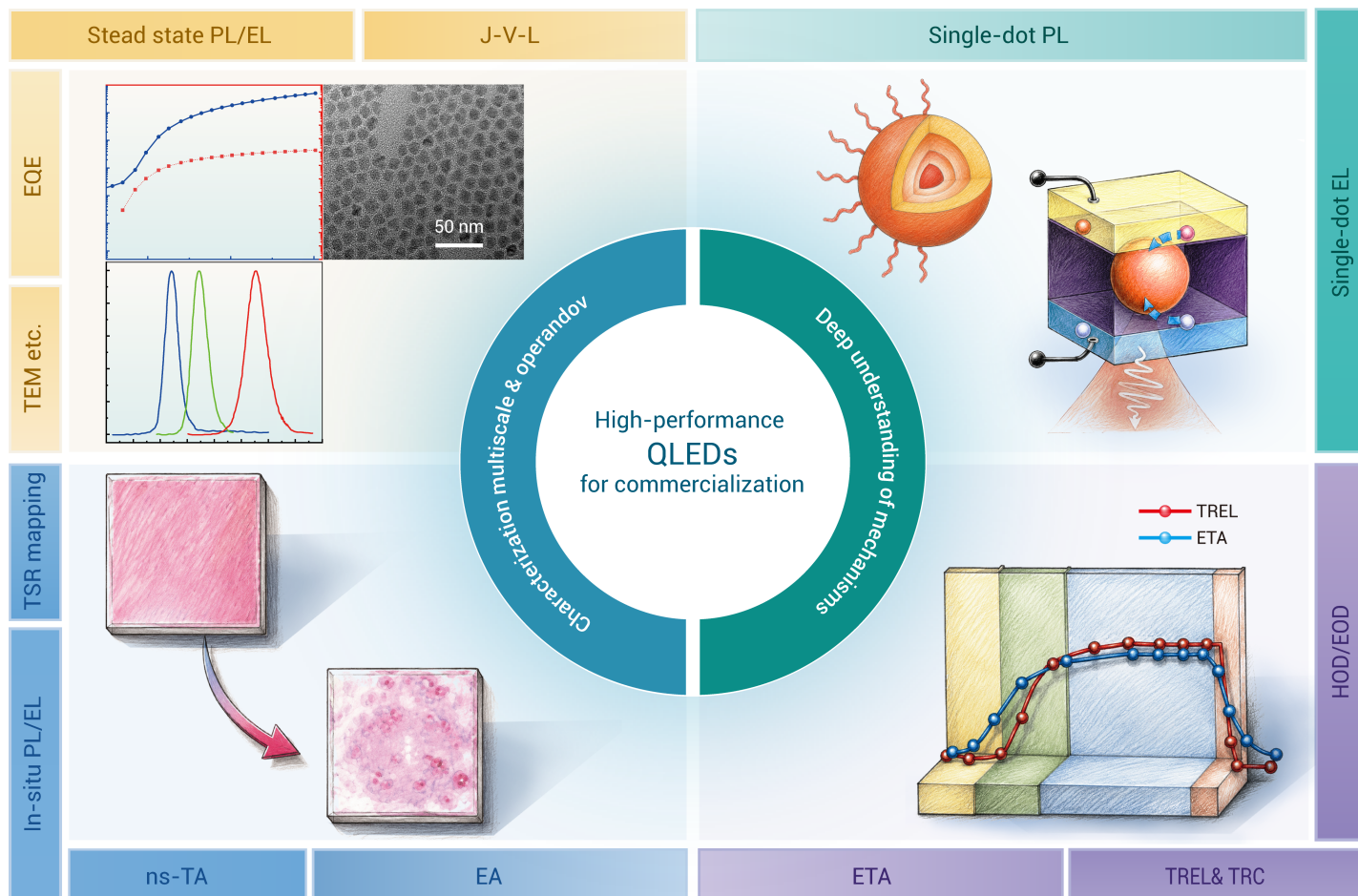
Yang Zhao,^{1,2} Boning Wu,^{3,*} Bo Li,⁴ Yaxuan Liu,^{1,2} Olga Kulalovich,⁵ Roman Vasiliev,⁶ Xingliang Dai,⁷ Wenming Tian,³ Fengjia Fan,^{4,8,9} Shengye Jin,³ and Shuai Chang^{2,*}

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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- Advanced operando and multiscale characterization of Quantum-dot light emitting diodes (QLEDs).
- Investigation of carrier dynamics, charge imbalance, interfacial interactions, and QLED degradation.
- Future directions and challenges toward highly efficient and stable QLED technologies.

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Quantum-dot light-emitting diodes (QLEDs) have great potential to deliver next-generation displays and lighting, yet it is difficult to achieve improvements in external quantum efficiency (EQE), roll-off, and reasonably operational stability due to their coupled process, such as charging-injection imbalance, carrier accumulation/leakage, field-induced effects, interfacial quenching, and irreversible degradation, which are hard to isolate with conventional steady-state measurement. The techniques of multiscale and operando characterization methods are summarized in this review and arranged in a hierarchical manner of diagnostic tools connecting observables to quantitative parameters and design rules applicable to devices. Single-dot photoluminescence (PL)/electroluminescence (EL) with photon-correlation and time-resolved detection discloses the intermittency during charging, the Auger loss, and charge-state signal injection/recombination rates at the bias. For the device level, injection delay is measured with single carrier structures and transient electroluminescence/current (TREL/TRC) measurement systems, and electrically excited transient absorption (ETA) is employed to inseparably isolate the carrier distributions of charge storage and recombination and leakage of quantum-confined Stark effect (QCSE). Additionally, electroabsorption (EA) tracks layer-resolved internal-field evolution and voltage-loss pathways during aging. The approaches used to differentiate between interfacial quenching and permanent damage, and localize the hotspots driven failure are complementary: nanosecond transient absorption (ns-TA), in-situ EL/PL, and time-gated mapping. These advanced characterization methods provide a deeper understanding of working mechanisms, enabling the development of more effective strategies to enhance the efficiency, stability, and long-term performance of QLEDs in practical applications.

INTRODUCTION

Quantum dot lighting-emitting diodes (QLEDs) have been of specific interest in recent decades for their potential applications of millions of displays and lighting in the next generation.¹⁻⁷ Having exceptionally impressive strength, such as purity of colors, extensive color gamut, and the ability to tune emitted wavelength,⁸⁻¹⁰ QLEDs are commonly thought to be the close rivals that may overcome the current display standards and enter entirely new areas of applications.¹¹⁻¹⁴ They consist of injection-type optoelectronic structures that include cathode, electron transport layer (ETL), quantum dot (QD) emission layer (EML), hole transport layer (HTL), hole injection layer (HIL), and anode.¹⁵ Their work can be generally described as consisting of four stages:¹⁶ (i) charge injection, where the electrons of the cathode overcome the interfacial barrier and are injected into ETL, and the anode onto holes into the HIL, after overcoming the anode-only HIL barrier; (ii) charge transport, where the injected carriers migrate through their respective transport layers toward the QD EML; (iii) recombination, once electrons and holes meet in the EML, they are bound to form excitons, which recombine radia-

tively to emit photons; (iv) emission and extraction, whereby the photons are extracted from the devices through the transparent electrode with efficiency influenced by optical losses.^{17,18} In practice, these processes are strongly influenced by the energy-level alignment across the multilayer stack (ETL/EML/HTL/HIL). Electron injection is typically more efficient than hole injection because of lower injection barrier and higher mobility in ETLs (e.g. ZnO), which leads to charge imbalance and electron accumulation in the EMLs.^{10,13,19} This imbalance, in turn, induces nonradiative losses and limits device performance.²⁰ Among the major parameters, the external quantum efficiency (EQE) and operational lifetime^{21,22} are the crucial factors to be considered when assessing the performance of the QLEDs.²³⁻²⁵

Since the first demonstration of QLEDs in the early 1990s,²⁶ QLEDs have indicated impressive improvements.²⁷⁻³² Owing to intensive efforts in materials optimization and device engineering, the maximum EQE of the three primary colors - red, green, blue- has been brought to over 20%, with values of 35.6%,²⁸ 28.7%,³¹ and 24.7%,³² respectively. These efficiencies, coupled with substantial gains in operational lifetimes - another crucial parameter for practical applications³ - have enabled the QLEDs to become increasingly more feasible in a wide variety of commercial applications. Notably, the T₉₅ operating lifetimes (defined as the time required for brightness to decrease to 95 % of its initial value at 1,000 cd/m²) of the three primary color QLEDs are 61,180 hours for red,²⁷ 17,700 hours for green²⁹ and 227 hours for blue.³³ Despite the rapid progress in QLEDs, several critical challenges remain, including efficiency roll-off at high brightness, charge imbalance, and limited operational stability, which significantly hinder their commercialization. Addressing these issues requires a deeper understanding of underlying physical processes related to carrier dynamics, exciton recombination, and device degradation. However, fundamental characterization techniques are often insufficient to resolve these coupled and transient processes.

Therefore, advanced characterization techniques are becoming increasing essential for uncovering the working mechanisms of QLEDs and enabling rational device optimization. Basic electrical/optical characterizations are employed in benchmarking and optimization of QLEDs, including *J-V-L* measurements, steady-state photoluminescence (PL) and electroluminescence (EL), efficiency, and lifetime tests.³⁴⁻³⁷ Nevertheless, such measures cannot explain the microscopic process³⁸ in terms of exciton recombination, charge accumulation, leakage, internal-field effects, and aging, thus it is not easy to trace individual loss mechanisms.³⁹ This review is hence concerned with new operando and multiscale characterization techniques that directly project observables in experiments on the physics of devices. Specifically, we systematize a hierarchical diagnostic toolbox across emitter limits (single-dot PL/EL), device-level charge/field dynamics (TREL/TRC, ETA, EOD/HOD), and aging & spatially heterogeneous degradation (EA, ns-TA, in situ EL/PL and time-gate mapping). By systematically decoupling competing mechanisms, these approaches can transform microscopic processes into practical design principles of confinement physics, injection balancing, interfacial passivation,

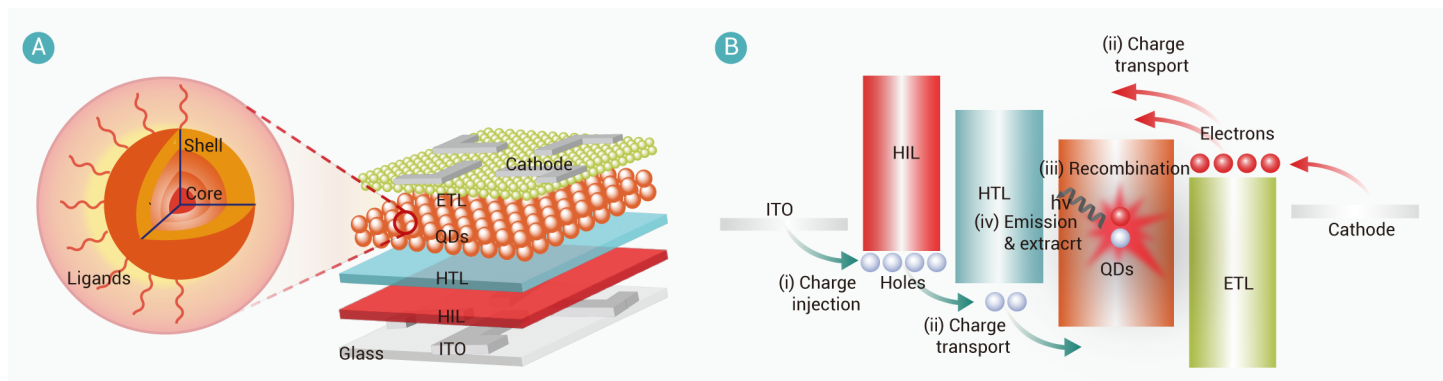


Figure 1. Structure and working mechanism of QLEDs (A) Schematic illustration of QLED structures and (B) Charge injection/transport and emission process in QLEDs.

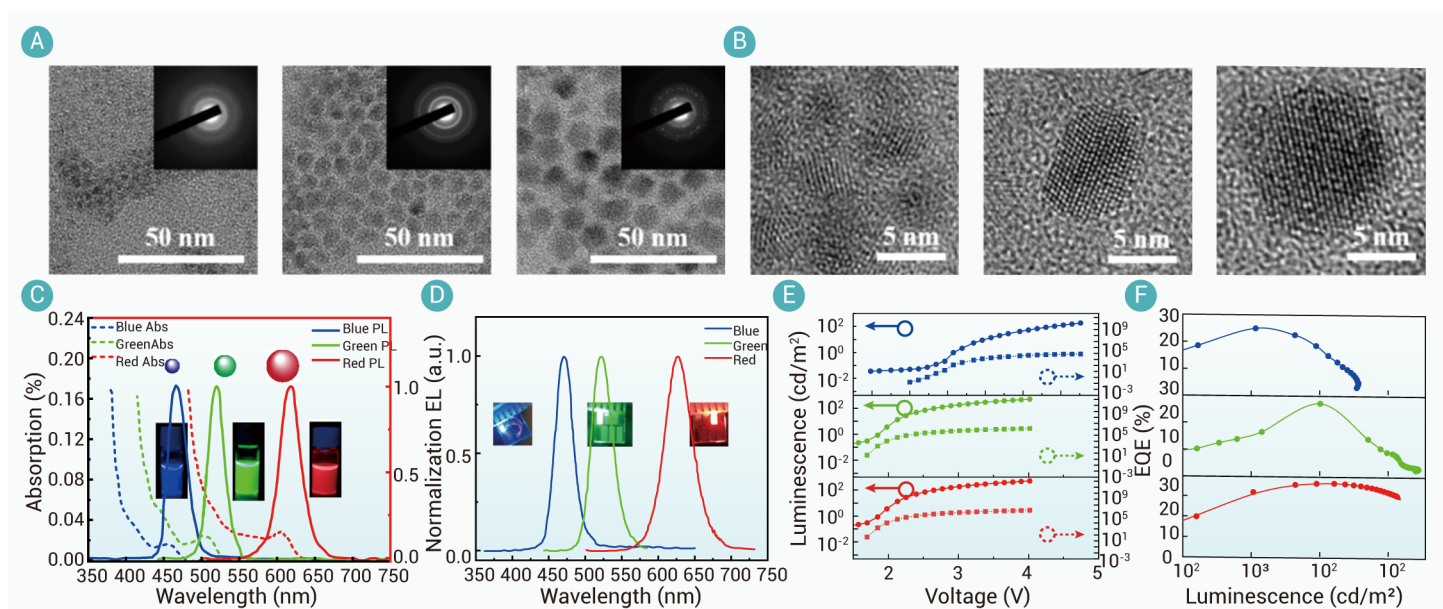


Figure 2. Structural, optical, and electroluminescent characterization of blue-, green-, and red-emitting QDs and corresponding QLEDs (A) TEM images of blue, green, and red QDs (left to right; insets: SEAD patterns).⁵³ (B) Corresponding HRTEM images (left to right). Reproduced under the terms of the CC-BY license.⁵³ Copyright 2015, Han et al., published by Optics Express. (C) UV-Vis absorption (dashed lines) and normalized PL spectra (solid lines) of blue, green, and red QDs (insets: photographs of QD solutions).⁵⁴ (D) Normalized EL spectra of blue, green, and red QLEDs (insets: photographs of QLED devices).⁵⁴ (E) J-V-L characteristics of blue (top),³² green (middle),⁵⁵ and red (bottom) QLEDs. (F) EQE-J characteristics of blue³² (top), green⁵⁵ (middle), and red²⁸ (bottom) QLEDs (Data extracted from these literatures.^{28,32,55}).

and stack engineering, thus enhancing the rapid rationalization of the efficiency and operational stability of QLEDs.

FUNDAMENTAL CHARACTERIZATION

Fundamental characterization plays a central role in evaluating QLED performance from materials to devices architectures.^{17,23} Quantum dots, serving as the core emissive components, are semiconductor nanocrystals with pronounced quantum confinement effects.^{40,41} When excited by optical or electrical energy, electrons are promoted to the conduction band, leaving behind holes in the valence band, thereby forming electron-hole pairs (excitons).^{42,43} Owing to nanoscale confinement,⁴⁴ the energy levels of QDs become discrete rather than continuous. After relaxation to lower energy states, electrons recombine radiatively with holes, releasing photons.⁴⁵⁻⁴⁹ The emission wavelength is determined by the size and composition of the QDs, resulting in narrow emission bandwidths, high color purity, and tunable emission.⁵⁰⁻⁵² To verify these structural and size-dependent optical properties, transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) were employed. As shown in Figure 2A, the TEM images of representative QDs with blue, green, and red QD samples were measured to be 2.47 ± 0.46 nm, 6.17 ± 1.23 nm, and 9.27 ± 0.95 nm, respectively.⁵³ These QDs exhibited uniform shapes and relatively narrow size distribution, as further confirmed by the selected-area electron diffraction patterns shown in the inset charts. Meanwhile, Figure 2B provided the corresponding HRTEM images, where

distinct lattice planes can be clearly observed, confirming the high crystallinity and well-defined crystal structures of the QDs.⁵³

To probe the optical properties, several basic characterization methods are employed. Spectroscopic techniques such as UV-Vis absorption and PL are used to evaluate band-edge transitions, emission spectra, full width at half maximum (FWHM), and quantum yields,⁵⁶⁻⁵⁹ thereby assessing color purity and radiative efficiency. From the absorption spectra of the blue, green, and red QDs shown in Figure 2C, the optical band gaps could be estimated as 2.66, 2.39, and 2.00 eV, respectively.⁵⁴ Figure 2C shows the normalized PL spectra of the corresponding QDs in solution, with maxima at 465 nm (blue), 518 nm (green), and 618 nm (red), and corresponding FWHM values of 26, 25, and 32 nm, respectively. At the device level, EL measurements provided direct insight into the optical output of QLEDs. As shown in Figure 2D, representative blue, green, and red devices exhibited EL peaks at 470, 522, and 622 nm, respectively.⁵⁴ Compared with the corresponding PL spectra, the EL spectra are slightly broadened (≈ 5 nm) and red-shifted, which can be attributed to strong interparticle interactions within the densely packed QD films as well as device heating under electrical bias.

Apart from spectral characteristics, Current density - voltage - luminance measurement established the fundamental relationship between charge injection, operating voltage, and brightness output.⁶⁰⁻⁶² From the *L-V* curve as shown in Figure 2E, the turn-on voltage (V_{on}) - defined as the voltage at which the luminance reaches 1 cd/m^2 - and the operating voltage at maximum

Table 1. Comparison between fundamental and advanced characterization techniques for QLEDs.

	Fundamental characterization	Advanced characterization
Measurement type	Steady-state electrical/optical measurement	Time-resolved and operando techniques
Temporal resolution	limited access to dynamics processes	High temporal resolution for transient dynamics
Information obtained	Macroscopic performance metrics	Microscopic processes (e.g., charge, exciton, fields)
Ability to decouple processes	Limited capability in decoupling coupled processes	Effective separation of coupled processes and identification of mechanisms
Operando capability	Limited under realistic operating conditions	Direct probing under realistic operation
Role in research	Benchmarking and evaluation	Mechanistic insight and device optimization guidance

luminance ($L_{\max}$). Together, these metrics quantify the device's driving requirements and provide a direct indication of its energy efficiency. As shown in Figure 2E, the V_{on} values of the blue, green, and red QLEDs were 3.4, 2.0, and 1.6 V, respectively.⁶³ To further assess how efficiently injected charges are converted into light, the EQE, defined as the ratio of emitted photons to the number of injected electron-hole pairs are evaluated.⁶⁴ The corresponding EQE-current density (EQE- J) characteristics for the blue, green, and red QLEDs are presented in Figure 2F. The EQE initially increases with luminance, reaches peak values of approximately 25%, 28%, and 35%, respectively, and subsequently decreases at higher luminance, reflecting a typical efficiency roll-off behavior associated with increased nonradiative recombination and charge imbalance.⁶⁵

In addition, key performance metrics – such as current efficiency (CE),⁶⁶ power efficiency (PE)⁶⁷ and chromaticity coordinates⁶⁸ etc. – could be extracted to enable a comprehensive evaluation of QLED performance.⁶⁹ Equally important for practical application is operational stability.⁷⁰ Lifetime testing is commonly performed by monitoring luminance evolution under constant current density driving, and the operational lifetime (e.g., T_{95} and T_{50}) is defined as the time when the relative luminance decays to 95% and 50% of the initial luminance, respectively.⁷¹ Among these metrics, EQE and operational lifetime are especially decisive, because together they largely determine the commercial viability of QLED technologies.⁸

Despite the value of these standard measurements in benchmarking devices and guiding material and structural optimization, they primarily deliver macroscopic figures of merit rather than mechanistic insight.⁶⁵ While parameters such as efficiency, operating voltage, and lifetime are readily obtained, the microscopic processes that govern efficiency roll-off and long-term degradation remain difficult to resolve.⁷² Critical phenomena – including exciton generation and recombination, interfacial charge transfer, trap formation, and chemical instabilities within functional layer – cannot be fully captured by conventional methods.⁷³ Consequently, advanced characterization techniques are required to directly probe emission mechanisms, carrier dynamics, and degradation pathways, thereby linking devices performance to the underlying physical processes more quantitatively and convincingly. To better illustrate the advantages of advanced characterization techniques, a comparison with fundamental methods is summarized in Table 1.

ADVANCED CHARACTERIZATION

Advanced characterization refers to a set of techniques that probe the dynamics and microscopic process in operating QLEDs. While fundamental characterization methods, such as steady-state electrical and optical measurements, provide essential macroscopic performance metrics, they fall short in revealing the intricate, dynamic processes governing QLED operation.¹³ These traditional techniques lack the temporal resolution to track ultrafast carrier dynamics, the spatial resolution to resolve local heterogeneity, and the capability to decouple simultaneously occurring microscopic mechanisms.⁷⁴ In this section, we explore how advanced, multi-scale in-situ characterization techniques address these limitations, offering deeper insights into QLED working mechanisms and guiding future device optimization.

Single-emitter physics

Single-dot photoluminescence. QDs exhibit superior optical properties

compared with conventional light-emitting materials; however, they often suffer from undesirable random intermittency in PL at the single-dot level, known as PL blinking.⁷⁵ Representative single-dot PL trajectories (Figure 3A) were first reported by Nirmal et al.⁷⁶ in 1996 using single-dot spectroscopy. PL blinking manifested as random switching between distinct “on” (bright) and “off” (dark) states under continuous excitation, a phenomenon invisible in conventional ensemble measurements. The statistics of “on”/“off” duration (Figures 3B–C) follow nonexponential, truncated power-law distributions. Extensive studies have attributed this intermittency to charge trapping and subsequent Auger recombination: once a carrier is trapped, the QD becomes charged and exciton recombination proceeds predominantly through nonradiative Auger pathways, resulting in the “off” state; when the trapped carrier is released, radiative recombination resumes, restoring the “on” state. Such instability represents a fundamental challenge for luminescent applications, as continuous and stable photo emission is essential for high-performance QLEDs.

Structural control has proven particularly effective in mitigating intermittency. Whereas wurtzite CdSe/CdS QDs typically require ultra-thick shells to achieve near-continuous emission, Qin et al.⁷⁷ demonstrated that phase-pure zinc-blende CdSe/CdS QDs exhibited strongly suppressed blinking ($\geq 95\%$ “on” time) even with shells of 4–16 monolayers (Figures 3D–E). Their emission statistics display markedly reduced off-time probabilities (Figure 3F), supported by stable antibunching signatures. These results highlighted the importance of symmetric crystal fields and well-confined carrier wavefunctions in minimizing access to trap and Auger-active regions.

Subsequent work has shown that PL blinking is closely tied to the charge state of QDs. Using single-dot spectroscopy (experimental setup in Figure 3G), Xu et al.⁷⁸ directly resolved neutral M_0 , negative M^- , and positive M^+ excitons, allowing precise measurement of their absorption cross sections (ACSS) and quantum efficiencies (QEs). They found that the presence of an extra carrier accelerates Auger recombination, significantly reducing radiative efficiency. Positive trions, in particular, exhibited faster Auger rates and lower QEs, due to stronger hole confinement and enhanced Coulomb interactions. Except that, charging altered PL saturation behavior and reduced the effective absorption cross section, linking ionization processes to exciton bleaching and blinking.

Moreover, charging was shown to modify PL saturation behavior and reduce ACS values, effectively linking ionization process to exciton bleaching. These results demonstrate that blinking fundamentally arises from random charging and discharging, underscoring the need to control charge dynamics at the single-dot level in addition to structural optimization. Using the mean photon counts of each emissive channel (M), they assigned charge states and calculate $g^{(2)}(\tau)$ from photon-detection events indicated by the shaded bands as show in Figure 3H. The central-to-side peak area ratios increased from 0.115 (M^0) to 0.250 (M^-) and 0.496 (M^+), indicating progressively weakened antibunching upon charging. Consistently, power-dependent measurements (Figure 3I) showed reduced PL intensity and earlier saturation for charged states, corroborating Auger-dominated recombination as the key link between stochastic charging and blinking. Together, these findings indicated that PL blinking fundamentally originates from stochastic charging and discharging process at the single-dot level.⁷⁹ Consequently, achieving suppressed or eliminated blinking required not only structural optimization – such as core/shell engineering and control of crystal phase and symmetry –

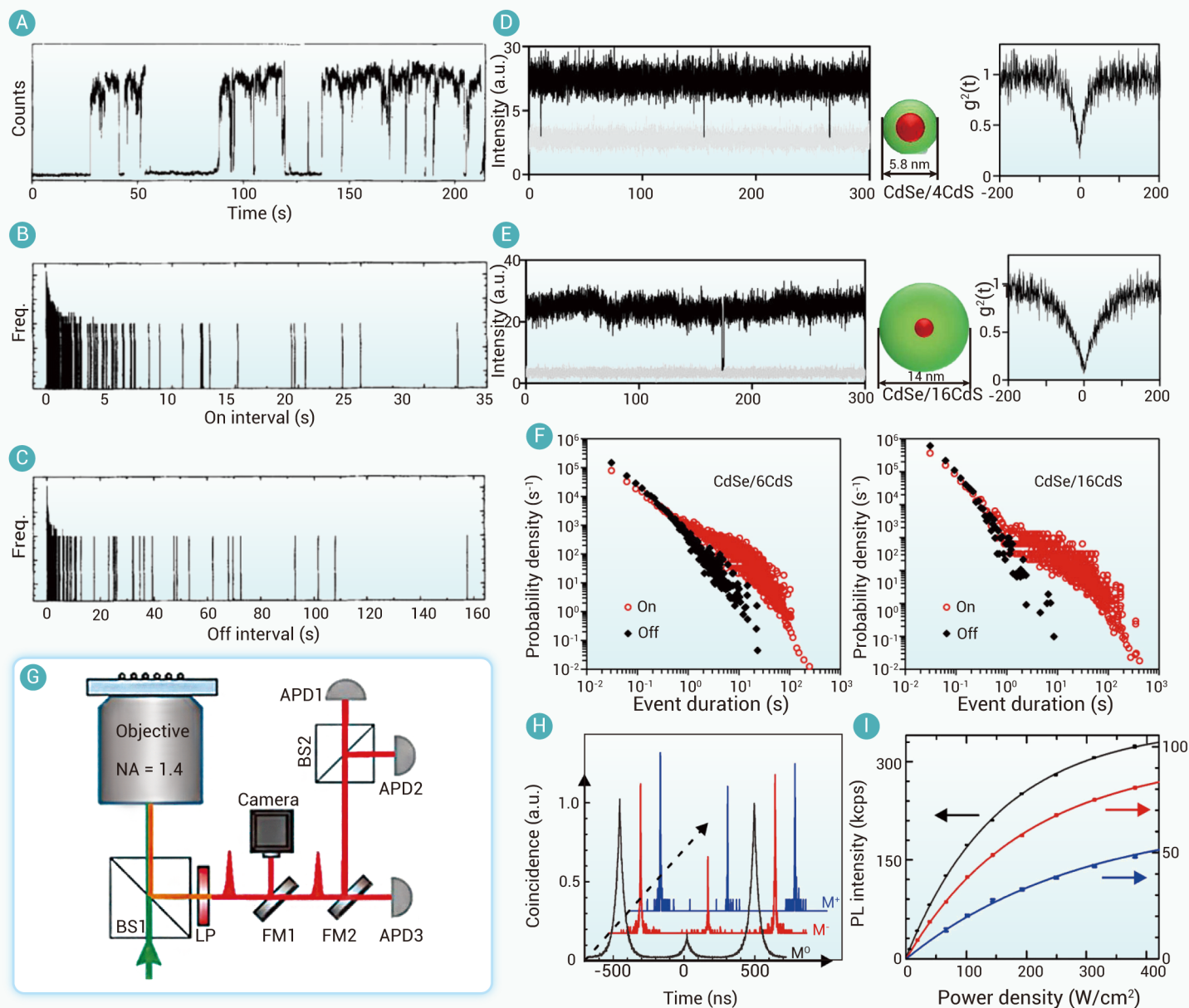


Figure 3. Single-dot blinking statistics and photo correction measurements of CdSe-Based QDs (A) PL intensity trajectory of a single CdSe nanocrystal (radius ≈ 2.1 nm).^{75,79} (B, C) Histograms of "on" and "off" time intervals for a single robust CdSe/ZnS core/shell nanocrystal recorded for ~ 39 min. Reproduced with permission.⁷⁹ Copyright 2017, Wiley. (D, E) Representative PL time traces of single zinc-blende CdSe/CdS core/shell QDs with a core radius of 1.55 nm and shell thicknesses of 1.36 and 5.44 nm corresponding to 4 and 16 monolayers.⁷⁷ (F) Log-log plots of probability density distributions of "on" (left) / "off" (right) for CdSe/CdS QDs with different shell thicknesses. Reproduced with permission.⁷⁷ Copyright 2013, American Chemical Society. (G) Schematic of the experimental setup (BS1:70:30; BS2: beamsplitter (50:50); LP: long-pass filter; FM: flap mirror; APD1/APD2 for $g^2(t)$ and PL time trace; APD3 for PL decay).⁷⁶ (H) $g^2(t)$ curves for the neutral state (M^0) and two charged states (M^- and M^+) at higher excitation.⁷⁶ (I) Saturation curves of PL intensity for M^0 (black), M^- (red), and M^+ (blue). Reproduced with permission.⁷⁶ Copyright 2017, American Chemical Society.

but also precise regulation of carrier ionization, trapping, and detrapping dynamics within individual quantum dots.

Single-dot PL spectroscopy serves as a fundamental platform for linking emission dynamics to charge-state evolution in quantum dots, providing unique insight into carrier trapping, ionization, and nonradiative recombination processes. However, its stochastic nature and limited representativeness restrict direct translation to device behavior. These limitations highlight the need for high-throughput and operando approaches to bridge single-emitter physics with practical QLED performance.

Single-dot electroluminescence. Although single-dot PL spectroscopy could probe the dynamics of photoexcited states, QLEDs operate under electrical bias, in which excitons are generated by injected charge carriers. Lin et al.⁸⁰ demonstrated electrically driven single-dot spectroscopy using a device in which isolated CdSe/CdS QDs are dispersed in an insulating PMMA matrix and placed between electron- and hole-transport layers, as shown in Figure 4A;

this configuration suppresses transport-layer electroluminescence and enables isolation of single-QD EL. Under electrical injection, emission can proceed through an expanded set of states, including neutral excitons $|X\rangle$, biexcitons $|BX\rangle$, and multiple charged excitonic configurations, reflecting injection-assisted charging and discharging (Figure 4B). Consistent with this mechanism, the EL shows systematically lower $g^2(0)$ values than PL across dots, reaching ~ 0.045 (Figure 4C), indicating enhanced single-photon purity under device-relevant operation. Overall, electrically pumped single dot spectroscopy provided a direct route to quantify charged dynamics and recombination pathways that were otherwise inaccessible under purely excitation.

Building on electrically pumped single-dot spectroscopy, Deng et al.⁸¹ further elucidated carrier-injection dynamics in CdSe/CdZnS QDs under device operation. By combining EL with time-resolved PL measurement (Figure 4D), they separated the slow, stochastic formation of electrically generated excitons (10^1 – 10^3 μ s) from their nanosecond radiative decay. The

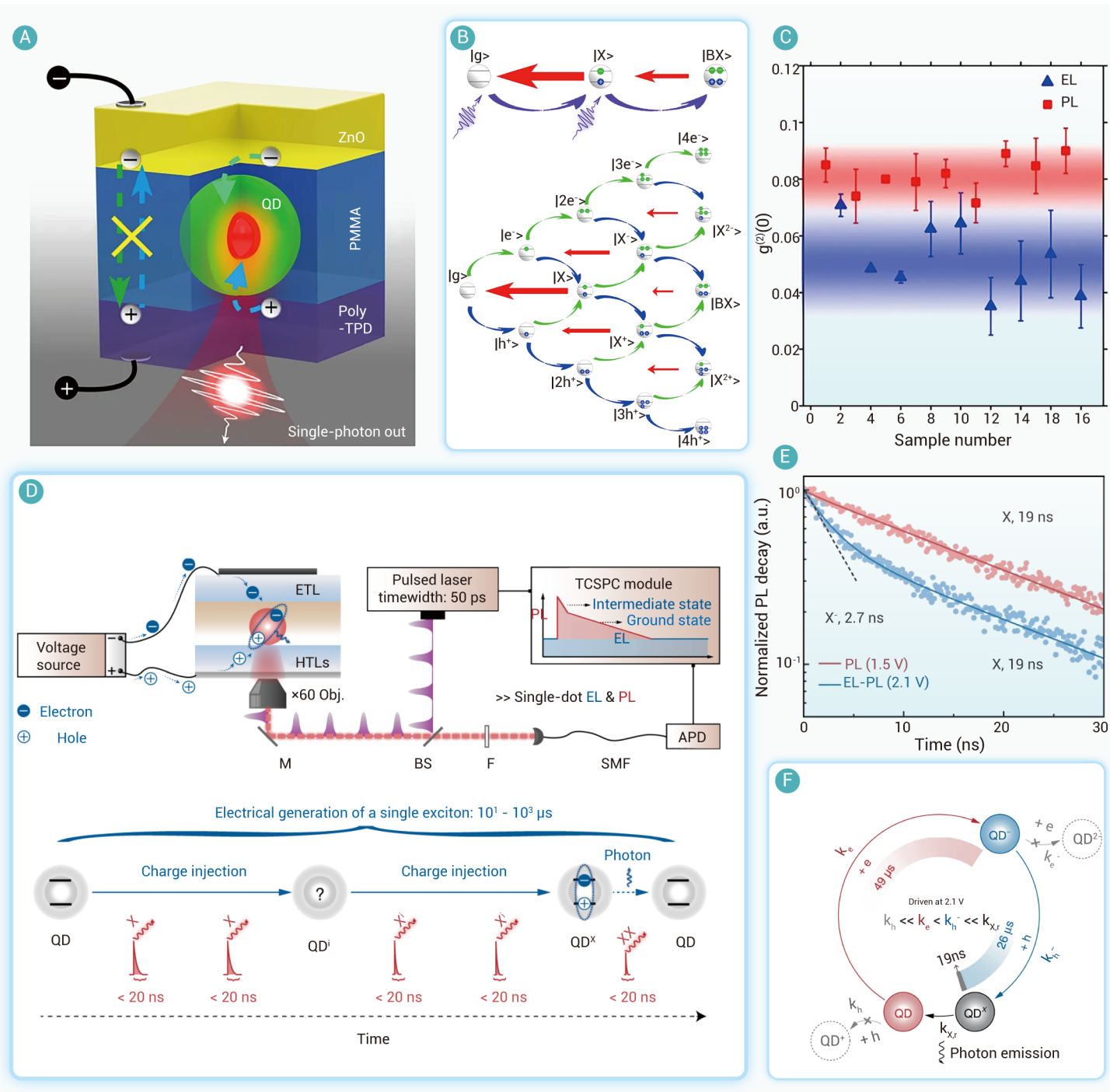


Figure 4. Single-dot EL mechanism and single-photon performance of QD devices (A) Devices schematic (ZnO/PMMA/Poly-TPD) enabling single-photon emission.⁸⁰ (B) State diagram of carrier charging and recombination processes involving the ground state $|g\rangle$, single-electron/hole states $|e\rangle$ and $|h\rangle$, multi-electron/hole states $|ne\rangle$ and $|nh\rangle$, neutral exciton $|X\rangle$, charged excitons $|X^{\pm}\rangle$ and $|X^{2\pm}\rangle$, higher-order charged states (e.g., $|X^{2\pm}\rangle$, $|X^{2\pm}\rangle$), and biexciton $|BX\rangle$, illustrating sequential carrier injection and radiative decay pathways.⁸⁰ (C) $g^{(2)}(0)$ values measured from 9 QDs under EL (blue) and PL (red); error bars denote the standard error from four points around zero delay. Reproduced under the terms of the CC-BY license.⁸⁰ Copyright 2017, Lin et al., published by Nature Communications. (D) Schematic carrier dynamics of single-dot EL and combined electrical injection and weak pulsed optical probing setup.⁸¹ (E) PL decay without injection (PL, 1.5 V: $\tau_X \approx 19$ ns) and with injection (EL-PL, 2.1 V: $\tau_X^- \approx 2.7$ ns) (G) Kinetic model of the EL cycle at 2.1 V with electron injection (EL-PL, 2.1 V: $\tau_X^- \approx 2.7$ ns).⁸¹ (F) Kinetic model of the EL cycle at 2.1 V with electron injection (red arrow, $k_e \approx 2.0 \times 10^4 s^{-1}$), hole injection (blue arrow, $k_h \approx 3.8 \times 10^4 s^{-1}$), and radiative recombination (black arrow, $k_{x,r} = 1/19 ns^{-1}$). Reproduced under the terms of the CC-BY license.⁸¹ Copyright 2020, Deng et al., published by Nature Communication.

time-resolved data revealed a fast decay component (~ 2.7 ns) associated with a negatively charged intermediate state (QD⁻), in contrast to the longer-lived neutral exciton (~ 19 ns), as illustrated in Figure 4E. The results indicated that electrical operation preferentially drove the QD into a negatively charged state, thereby facilitating subsequent hole injection and enhancing exciton formation efficiency (Figure 4F). Together with the observed spectral broadening and red-shift under bias, this study highlighted charge accumulation and injection asymmetry as key factors governing exciton dynamics and

emission behavior in operating QLEDs. These findings demonstrate that EL single-dot spectroscopy is an essential tool for understanding carrier injection, recombination, and exciton dynamics in QD-based EL devices. By enabling direct observation of the electrical excitation, this technique provides critical insights for optimizing charge injection efficiency and improving overall device performance.

Single-dot EL spectroscopy serves as a fundamental platform for linking carrier injection dynamics to charge-state evolution in quantum dots under

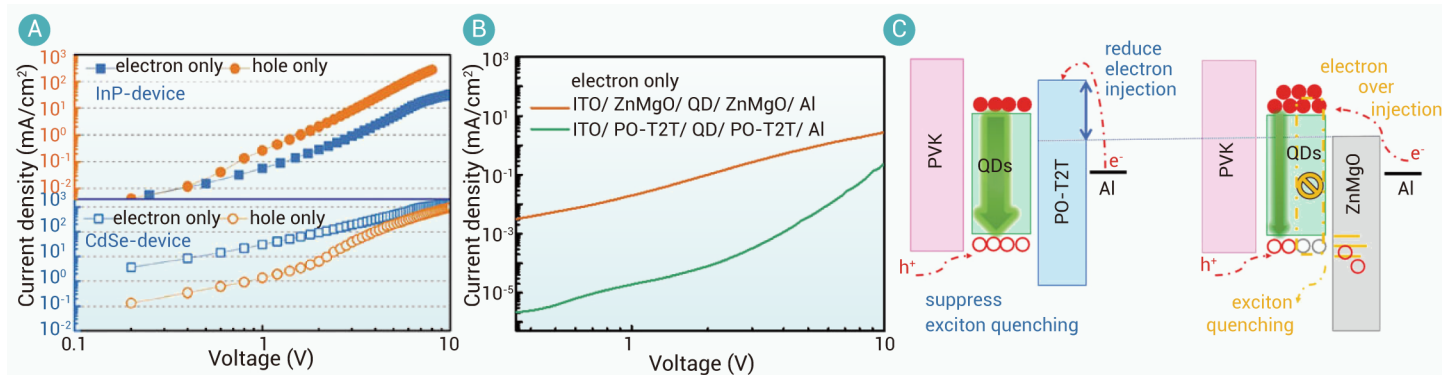


Figure 5. Device performance and electron injection characteristics of InP- and CdSe-QLEDs (A) J–V characteristics of the hole-only and electron-only devices for InP- and CdSe-QLEDs. Reproduced under the terms of the CC-BY license.⁸⁵ Copyright 2024, Su et al., published by Advanced Physics Research. (B) J–V characteristics of electron-only devices with different ETLs (ITO/ZnMgO/QD/Al and ITO/PO-T2T/QD/Al).⁸⁵ (C) Schematic of electron injection with different ETLs. Replacing ZnMgO with PO-T2T improves charge balance and suppresses exciton quenching due to the lower defect density of PO-T2T. Reproduced with permission.⁸⁹ Copyright 2022, Wiley.

operation, providing unique insight into injection-induced charging and recombination pathways. However, its reliance on complex device architectures and limited throughput restricts its applicability and statistical representativeness. These limitations highlight the need for high-throughput and operando approaches to bridge single-emitter insights with practical device performance.

It should be noted that key phenomenon revealed by single-dot PL and EL, such as blinking, Auger recombination, and charging-induced emission fluctuations, are generally observed across Cd-based, InP-based, and other QDs.⁸² However, their specific characteristics depend strongly on material composition and surface passivation. Nevertheless, measurements on individual dots may not represent ensemble or device behavior because of stochastic variability.^{83,84} In particular, single-dot EL usually requires specialized architecture with low signal throughput. Future efforts should focus on high-throughput and operando approaches to better bridge single-emitter physics with device performance.

Operando charge dynamics

Single-carrier devices. Single-carrier devices, categorized into hole-only devices (HODs) and electron-only devices (EODs), are effective for disentangling carrier injection and transport in QLED stacks.^{85,86} As shown in Figure 5A, Su et al.^{87,88} found that InP-based structures exhibited higher hole-only currents than electron-only currents, indicating a pronounced hole-injection advantage and potential hole over-injection in the full devices. Capacitance-voltage and luminance measurements further support this assignment: incorporating a hole-blocking configuration reduced the capacitance feature attributed to hole accumulation and altered the luminance evolution in InP-QLEDs. In contrast, CdSe-QLEDs showed increased capacitance upon hole blocking, consistent with uncompensated electron accumulation and thus hole-limited operation. These results underscore that charge-balance engineering is emitter-dependent, requiring mitigation of excess holes injection in InP-based QLEDs through suppressed hole injection or enhanced electron injection, but enhanced hole injection/transport in CdSe-based devices.

Leveraging this insight, Gao et al.⁸⁹ tuned electron injection in InP-QLEDs by replacing ZnMgO with an organic molecule of 2,4,6-tris(3-(diphenylphosphoryl)phenyl)-1,3,5-triazine (PO-T2T), which substantially lowered the electron-only current (Figure 5B) but paradoxically improved luminance and EQE. The schematic in Figure 5C attributed this enhancement to suppressed electron over injection, reduced interfacial charge accumulation, and mitigated exciton quenching, underscoring that charge-management strategies must be tailored to the emitter system.

Building on the above conclusion that InP-QLEDs benefit from moderated electron injection to restore charge balance, tri-layer p-i-n architectures offered an integrated route to tune injection and suppress leakage.⁹⁰ HOD tests confirmed that Spiro-OMeTAD: TFB BHJ HTL enhanced hole injection compared to PEDOT: PSS/TFB, as indicated by higher current density and lower capacitance. EOD tests revealed that using eutectic gallium indium (EGaIn) as the cathode reduced electron injection and leakage, improving

stability and efficiency, with a peak EQE of 27.3%. these results demonstrate the critical role of efficient charge injection in achieving better performance and stability in QLEDs. For comparison, HODs were also fabricated, and their tests confirmed that the improvement in performance was not due to changes in hole injection. This reinforces the conclusion that positive aging primarily results from enhanced electron injection. These findings highlight the critical role of managing electron injection to improve charge balance, efficiency, and long-term performance in QLEDs.

Single-carrier devices serve as a fundamental platform for disentangling carrier injection and transport processes in QLED stacks, proving insight into charge imbalance, injection asymmetry, and carrier accumulation. However, their simplified device configuration limits direct representation of full device operation and may overlook coupled carrier dynamics under realistic conditions. These limitations highlight the need for complementary operando approaches to bridge single-carrier analysis with practical QLED performance.

Transient electroluminescence. To directly capture carrier injection, accumulation, and recombination dynamics under realistic operation, transient electroluminescence (TREL) is widely employed in QLED studies.^{91–93} As shown schematically in Figure 6A, a periodic voltage pulse generated by a signal generator is applied to the devices through a rheostat, while the electroluminescence is collected by a high-speed photomultiplier tube (PMT) or APD and recorded together with a reference signal on an oscilloscope. A typical TREL spectroscopy could be roughly partitioned into four chronological regimes as show in Figure 6B. (I) Delay stage: EL is absent immediately after the pulse is applied. This delay mainly originates from the transient RC charging of the device and interfacial charge accumulation, as reflected by the fast component in the transient current (TRC). During this stage, the applied bias is primarily used to charge the device capacitance and establish the internal electric field. EL turns on only after sufficient charging enables effective carrier injection into EML. (II) Rising stage: EL turns on and increases as more electrons and holes begin to overlap in the EML and form excitons. (III) Steady (balance) stage: EL reaches a plateau, indicating a quasi-steady state where carrier injection and transport are balanced by recombination. (IV) Decay stage: after the pulse is switched off, EL decays as residual carriers/excitons recombine and stored charges are released.^{94,95} Importantly, the optical TREL signal is complemented by the time-resolved response, which directly reports the device's electrical charging/discharging under the same pulsed bias. In practice, the device response can be interpreted with an equivalent RC-type circuit, where a fast current component corresponds to capacitive charging and interfacial carrier accumulation, followed by a slower component associated with charge release and recombination. By jointly analyzing TREL and TRC, one can link the onset and growth of EL to carrier arrival and charge build-up, and correlate the EL plateau/decay with injection balance and post-pulse discharge processes. The carrier distribution evolution depicted in Figure 6C provides a physical picture for this correlation. During the rising stage (II), the progressive charging of the device and accumulation of the interfacial charges reshape the internal electrical field and

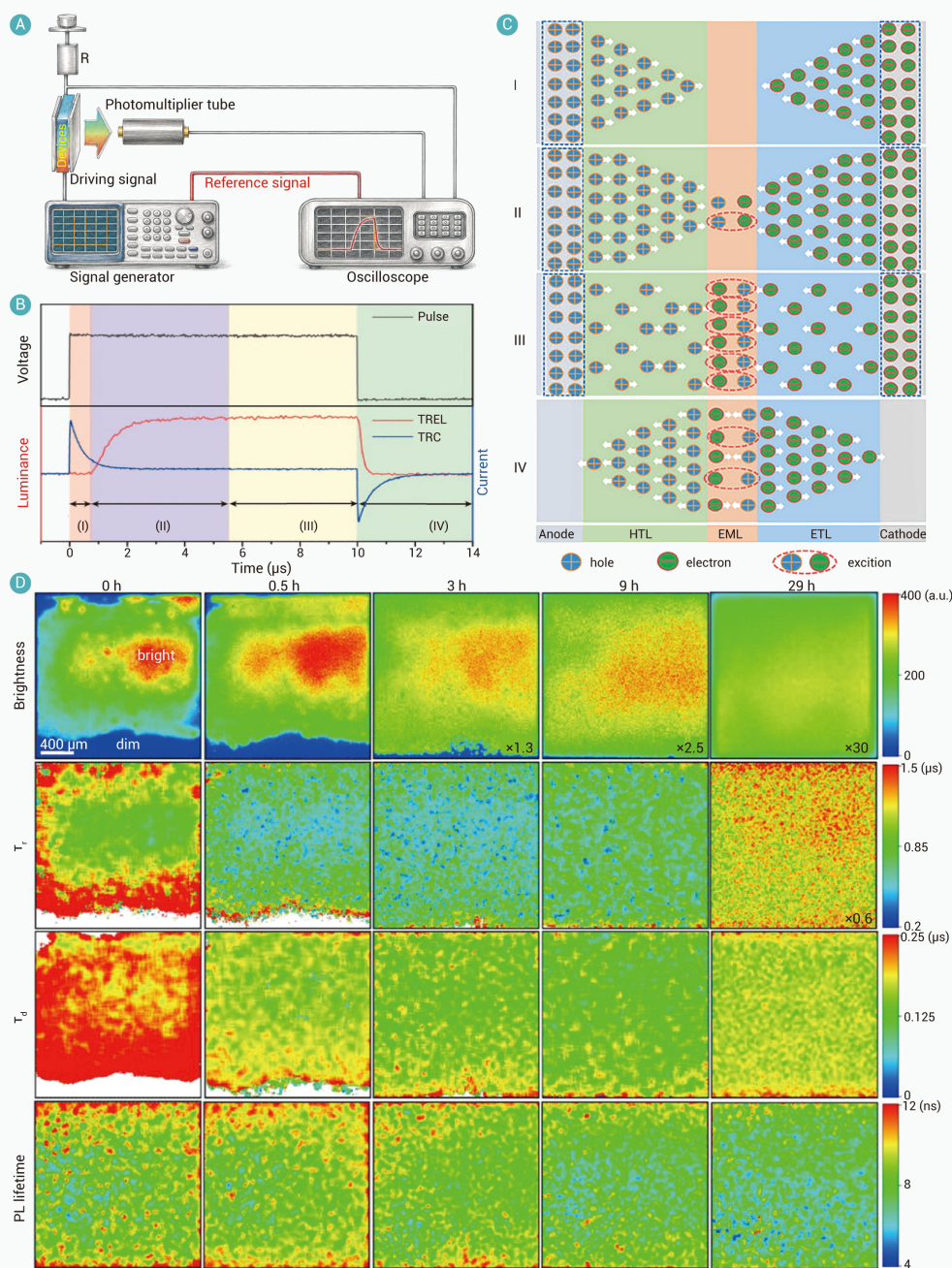


Figure 6. Measurement setup and degradation analysis of QDs in planar LEDs (A) Schematic diagram of test measurement setups. Reproduced with permission.⁹¹ Copyright 2024, Wiley (B) Typical TRC and TREL curve and its stage division. Period I: delay stage. Period II: rising stage. Period III: balance stage. Period IV: decay stage. Reproduced with permission.⁹⁴ Copyright 2023, American Chemical Society. (C) Schematic illustration of the charge distribution during the four stages of TREL response in a planar LED. Reproduced with permission.⁹¹ Copyright 2024, Wiley. (D) Equilibrium brightness, τ_r of TREL, τ_d of TREL, and exciton lifetime of the QDs after different hours of degradation (0, 0.5, 3, 9, 29 hours). Reproduced with permission.⁹⁶ Copyright 2025, Wiley.

ally dim, whereas initially dim regions brighten (positive degradation), leading to a more uniform emission pattern. The concurrent evolution of τ_r and τ_d indicated that these brightness changes are coupled to spatially varying injection/charge dynamics, highlighting local injection imbalance as key factor governing degradation. It is noteworthy that TREL mapping holds strong potential for extracting the spatial distribution of key temporal parameters, which could, in principle, be correlated with local carrier dynamics and defect distribution. Such analysis might further enable the identification of early-stage failure hotspots, providing valuable guidance for device optimization and improving the operating stability of QLEDs.

TREL resolves time dependent carrier injection, accumulation, and recombination dynamics in operating QLEDs, linking electrical charging process with light emission. When extended to spatial mapping, it further enables visualization of local injection heterogeneity and degradation evolution across the devices. However, its interpretation can be complicated by RC delay effects and coupled carrier dynamics, while the quantitative extraction of microscopic parameters often relies on model assumptions. These limitations highlight the need for integrated multi-modal and operando approaches to fully capture charge dynamics and degradation mechanisms in practical QLEDs.

lower the effective injection barriers, enabling increasing carrier injection into EML and the formation of an expanding recombination zone. As the devices approaches the steady stage (III), a quasi-equilibrium is established in which carrier injection, transport, and recombination are balanced, leading to a stable EL plateau. Once the driving field is removed (IV), the stored charges are released and residual carriers/excitons recombine, giving rise to the EL decay.⁹¹ Together, TREL-TRC measurements offer a time-domain framework to quality how charge injection and accumulation govern exciton formation and light emission in operating QLEDs.

Building on the time-domain TREL-TRC framework above, TREL mapping extended transient analysis into the spatial domain, allowing charge injection and degradation to be tracked locally across an operating QLED. In the implementation reported by Tian and co-worker,⁹⁶ the device area is imaged onto a high-resolution grid (256 × 256 pixels, ~ 2 × 2 μm per pixel), effectively treating each pixel as a “mini-QLED” from which both the local brightness and characteristic transient time constants - delay time (τ_d), rise time (τ_r), and decay time (τ_d) - could be extracted. As shown in Figure 6D, the brightness distribution redistributed during aging (0–29 h): initially bright regions gradu-

Electrically excited transient absorption spectroscopy. As illustrated in Figure 7A, electrically excited transient absorption (ETA) spectroscopy is an operando analogue of optically pumped transient absorption (TA).⁵⁵ In TA (Figure 7A, left), a femtosecond laser pulse optically excited the sample and a time-delayed supercontinuum white-light probe tracked the ensuing spectral evolution as a function of delay time Δt . In ETA (Figure 7A, right), the optical pump was replaced by an electrical pulse delivered from an arbitrary waveform generator, which drove carrier injection into a QLED under realistic bias conditions; the same white-light probe and delay scan then recorded the transient absorption response during device operation. It is noteworthy that ETA differs fundamentally from conventional TA in both excitation method and experimental configuration. In TA, carriers are generated via optical excitation, and the subsequent dynamics are probed under photoexcited conditions, while ETA could enable direct access to microscopic processes occurring in operating devices, including carrier injection, accumulation, transport and recombination, as well as electric-field-induced effects. Such processes are either inaccessible or inaccurately represented in conventional TA measurements. Consequently, ETA directly connected microscopic injection/

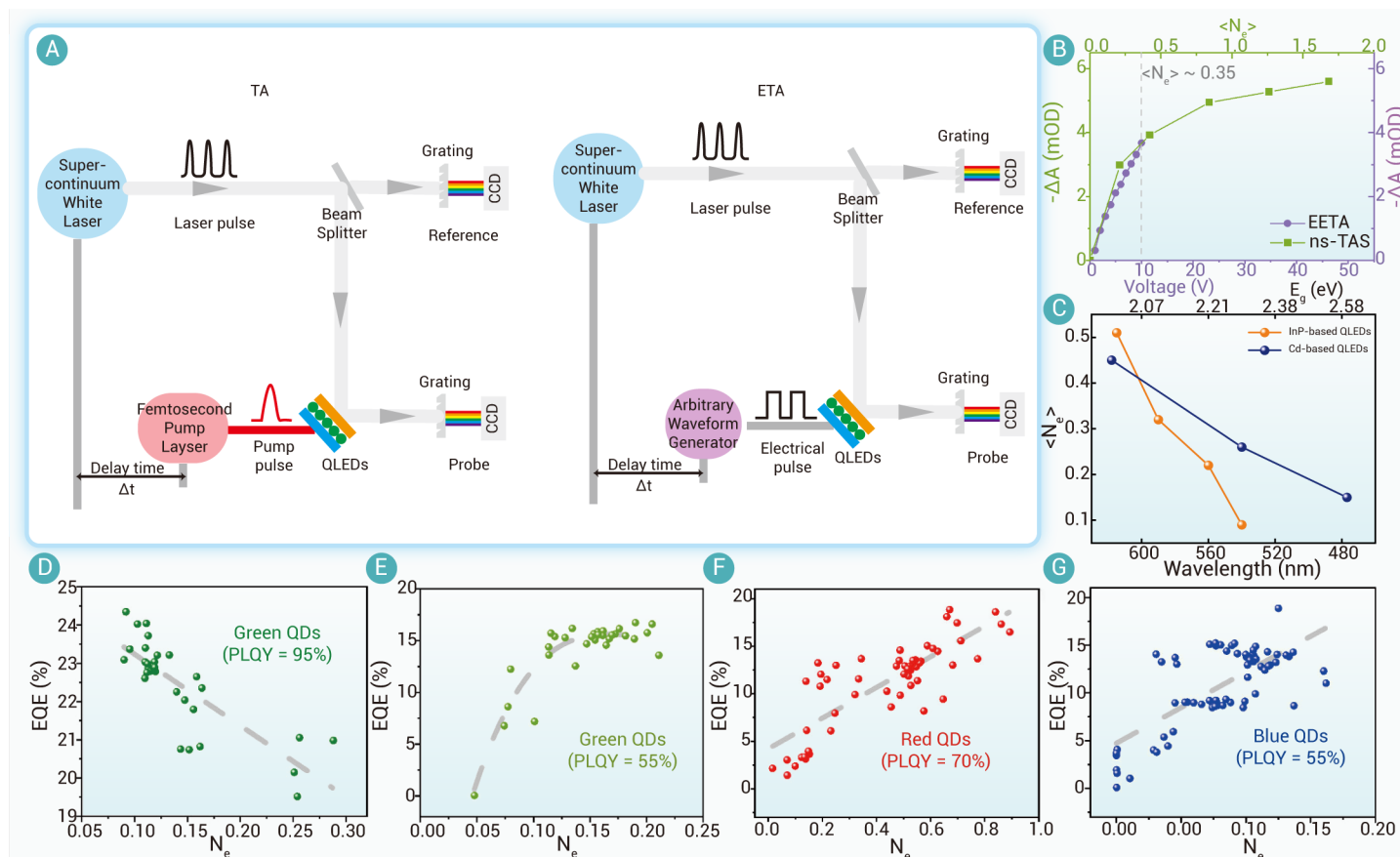


Figure 7. ETA setup and exciton population-efficiency relationship (A) Schematic of TA (left) and ETA (right) setups. (B) Comparison of band-edge state bleaching in ns-TAS (green) and ETA (purple). At 10 V, ETA bleaching corresponds to an average electron population of ~ 0.35 per QD in ns-TAS. Reproduced under the terms of the CC-BY license.¹⁰¹ Copyright 2024, Wang et al., published by Nature Communication. (C) Peak average electron population per QD for InP-based (615 nm, 593 nm, 560 nm, and 540 nm; orange squares) and Cd-based QLEDs (636 nm, 529 nm, and 473 nm; blue circles).⁵⁵ (D - G) Correction of electron population (N_e) and EQE for QLEDs with (D) green QDs (95% PLQY at 3.5V), (E) green QDs (55% PLQY at 3V), (F) red QDs (70% PLQY at 4V), and (G) blue QDs (55% PLQY at 6V). Reproduced under the terms of the CC-BY-NC-ND.¹⁰² Copyright 2024, Yan et al., published by Nano Letters.

charging dynamics with device-relevant electric-field effects that are inaccessible under purely photoexcited conditions. In ETA, the absorption change ΔA is defined as the absorbance difference between electrically pumped and unpumped states. To quantify the average electron population per quantum dot, $\langle N_e \rangle$, a calibration is first established from TA by correlating the band-edge (1S) bleach amplitude with the average photoexcited occupancy $\langle N \rangle$, where $\langle N \rangle = P\sigma$ (P is the excitation power density and σ is the absorption cross-section of the QD film determined from film absorbance and dot areal density). In operating devices, ΔA generally contains both carrier-induced bleaching and an electric-field-induced Stark component; the latter can be molded as a linear combination of the first and second derivatives of the steady-state absorption spectrum and subtracted to isolate the pure bleach amplitude. Mapping this electrically induced bleach onto the TA-derived calibration yields $\langle N_e \rangle$ as a function of bias,⁹⁷⁻¹⁰⁰ proving a quantitative operando metric for electron injection, accumulation, and leakage. Additionally, ETA also could provide critical insight into the origin of efficiency roll-off in QLEDs. The results revealed that the carrier population remained insufficient to induce significant multicarrier interactions, thereby ruling out Auger recombination as the dominant loss mechanism. In contrast, the evolution of bleaching and electric-field-induced Stark signals provided directly evidence of electron leakage. Specifically, the reduction of bleach amplitude, as with as the enhancement of Stark components at high bias, indicated a portion of electrons escaped from the emissive layer into adjacent layers. This process reduced the effective recombination probability and led to a decline in EQE.

Employing this approach, Gao et al.⁹⁷ extracted $\langle N_e \rangle$ and the electron quasi-Fermi level splitting E_{Fe} , establishing a charge-filling-dominated operating regime that contrasted with scenarios in which ground-state filling can be ruled out because $\langle N_e \rangle$ remained low. At 2 V, a monolayer device based on large full-composition-gradient QDs (FCG-QDs, ~ 18.4 nm) reaches $\langle N_e \rangle =$

0.23 (vs ~ 0.01 for conventional ~ 10 nm QDs), together with $E_{Fe} = 0.84$ – 0.92 eV at 2–3 V, confirming substantially enhanced per-dot charging filling under low bias. This filling-enabled strategy delivered $1,100$ cd/m^2 at 100% bandgap voltage and $6,000$ cd/m^2 at only 115% bandgap voltage while maintaining 21–23% PCE, minimal heating (0°C rise @ $1,000$ cd/m^2 for 2 h; $\sim 1^\circ\text{C}$ @ $6,000$ cd/m^2), and a T_{95} lifetime exceeding 48,000 h. Wang et al.¹⁰¹ quantified electron density in $\text{Cd}_x\text{Zn}_{1-x}$ Se-ZnS QLEDs and found $\langle N_e \rangle < 0.35$ even at 10 V (as illustrated in Figure 7B), excluding ground-state filling as the origin of enhanced high-energy light-hole (LH) electroluminescence; instead, field-induced suppression of the heavy-hole (HH) exciton oscillator strength was more pronounced than for LH excitons, rationalizing the spectral redistribution. Bian et al.⁵⁵ further showed that $\langle N_e \rangle$ decreased with shorter emission wavelength in Cd-based QLEDs (0.45 for red, 0.26 for green, 0.15 for blue), as shown in Figure 7C., accompanied by a drop in EQE (from 27.11% to 21.00%), and that conventional InP-ZnSeS-ZnS QLEDs could suffer from particularly low $\langle N_e \rangle$ in green emitters (0.09 vs 0.50 in red) (Figure 7C), leading to a sharp EQE loss (21.71% to 12.52%), consistent with trap states remaining unsaturated and non-radiative recombination dominating. Moreover, Yan et al.¹⁰² revealed a dual role of electron accumulation: as illustrated in Figures 7D–G, $\langle N_e \rangle$ correlated negatively with EQE in high-PLQY QDs (95%, green) (due to Auger/leakage), but positively in low-to-medium-PLQY QDs (55% green, 70% red, 70% blue) where electron accumulation facilitated trap filling and improved radiative recombination efficiency. This dual effect manifested as a transition from EQE enhancement at moderate electron accumulation to efficiency roll-off at excessive accumulation. These results indicated electron injection should be tailored to QD quality – restrained in high-PLQY systems to suppress nonradiative losses, but moderately enhanced in trap-rich QDs to improve carrier utilization, highlighting the importance of achieving a balance between trap passivation and suppression of nonradiative recombination.

Yan and co-authors¹⁰⁰ further employed ETA to resolve operando electron injection and release dynamics in QLEDs. By analyzing the ΔA in conjunction with TREL and TRC, as shown in Figure 8A, they extracted key kinetic parameters, including an electron injection delay of ~ 21 ns (red QLEDs), biexponential electron release (fast ~ 258 ns; slow > 1 μ s), and equilibrium electron population $\langle N_e \rangle \approx 0.62$ per dot. Meanwhile, the Stark component allowed the evolution of the internal field to be tracked, revealing a flat-band condition at ~ 210 ns. Importantly, electron injection was found to precede hole injection by ~ 233 ns, with electron-hole injection approaching a dynamic equilibrium at ~ 1 μ s, providing a quantitative timeline for charge-transport process relevant to efficiency and stability. Complementarily, Fan et al.¹⁰³ combined ETA with TREL to systematically compare electron-hole injection across red, green, and blue QLEDs at different temperatures ranging from 140 to 298 K. Electron injection exhibited weak temperature dependence ($\Delta \langle N_e \rangle < 10\%$) and followed a space-charge-limited-current (SCLC) behavior, whereas hole injection was strongly thermally activated (2 – 5 times increase) and produced a 0.3–0.5 μ s electroluminescence delay attributed to hole accumulation in the HTL (Figures 8B–C). As a consequence, the carrier imbalance and electron accumulation exhibit a strong dependence on temperature. The consistency of these trends across different HTLs, ETLs, and emission colors established a broadly applicable framework for diagnosing charge imbalance and guiding charge-balance engineering in QLEDs.

Li et al.¹⁰⁴ employed ETA to correlate the operando electron population, $\langle N_e \rangle$, with efficiency roll-off. At the onset of roll-off, $\langle N_e \rangle$ was only ~ 0.1 – 0.2 and remained low (~ 0.1 – 0.3) even at higher bias, far below the level required to trigger Auger recombination (~ 2 electrons per dot), thereby excluding Auger recombination as the dominant roll-off mechanism, as shown in Figure 8D. Comparing devices with different electron confinement further supported a leakage-driven picture: in CdSe/CdS QLEDs (weaker confinement), $\langle N_e \rangle$ plateaued beyond ~ 3 V and roll-off became pronounced, whereas CdZnSeS/ZnS devices (stronger confinement enabled by a higher-barrier ZnS shell) maintained a slowly increasing $\langle N_e \rangle$ and exhibited markedly suppressed roll-off. These results identified electron leakage as the primary origin of roll-off and directly motivate confinement engineering (e.g., thicker/higher-barrier ZnS shells) to mitigate it. Consistent conclusions were reported by Yan et al., who deconvoluted ΔA into bleaching, Stark (field-induced quenching), and leakage contributions. In a green QLED with peak EQE of 26.8%, leakage accounted for $\sim 95\%$ of the roll-off, while field-induced quenching contributed $\sim 5\%$ and Auger recombination was negligible, reinforcing leakage suppression as the key optimization target. Beyond efficiency, ETA has also been extended to diagnose operational stability. In NIR InAs-based QLEDs, Li et al.¹⁰⁵ tracked electric-field-induced Stark signatures to pinpoint aging origins: conventional devices degraded mainly via HTL (TFB) oxidation, evidenced by a $\sim 34\%$ of the TFB-related Stark signal (420 nm) with minimal change in the ZnSe-shell feature (445 nm), whereas in devices with a crosslinked HTL, the HTL Stark signal remained stable (393 nm) and degradation was instead associated with QDs (ZnSe-shell Stark signal decreased by $\sim 21\%$), as illustrated in Figures 8E–F. Collectively, these studies highlight ETA as a uniquely informative operando tool for quantitatively separating loss channels and guiding both roll-off mitigation and stability-oriented device design.

Another key capability of ETA is the operando quantification of the quantum-confined Stark effect (QCSE) in working QLEDs by tracking the electric-field-induced absorption changes (ΔA) and separating them from carrier-induced bleaching.¹⁰⁶ Under reverse bias, where carrier injection is negligible, ΔA is dominated by pure QCSE and can be used as a quantitative reference. Under forward bias, ΔA contains both bleaching and Stark components; these can be disentangled by fitting ΔA as a linear combination of a bleaching spectrum and derivative-shaped Stark response, and then benchmarking the extracted Stark amplitude against the reverse-bias calibration to obtain the effective QCSE strength under operation. Using this approach, a pronounced size dependence was revealed as shown in the Figures 8G: small QDs (~ 3.4 nm) are relatively QCSE-robust owing to strong confinement, large QDs (~ 6.5 nm) exhibit substantial field screening, whereas intermediate QDs (~ 4.2 nm) show the strongest QCSE response, with $\sim 38\%$ exciton quenching at +5 V forward bias – among the largest quenching levels reported under realistic operating conditions. Overall, these results position

ETA as a practical spectroscopic bridge between internal electric fields and device performance, and motivate strategies such as QD size selection and enhanced charge screening to suppress QCSE-induced exciton quenching and improving QLED efficiency and stability. ETA spectroscopy has emerged as a uniquely informative operando tool for QLEDs because it simultaneously quantifies carrier injection/accumulation and internal electric-field effects. By disentangling bleaching, Stark responses, and leakage-related features in ΔA , ETA provides direct, time-resolved metrics to diagnose key performance limits—such as charge imbalance, efficiency roll-off dominated by electron leakage, and QCSE-induced exciton timeline and its temperature dependence. These capabilities translate microscopic charge and field dynamics into actionable design rules (e.g., confinement engineering, injection control, and size selection), thereby enabling rational optimization of QLED efficiency, stability, and operational lifetime.

ETA spectroscopy enables quantitative probing of carrier injection, accumulation, and leakage dynamics in operating QLEDs, providing unique insight into interplay between charge population, electric-field effects, and exciton recombination processes. It further allows direct identification of dominant loss mechanisms, such as ruling out Auger recombination processes and confirming electron leakage as the primary origin of efficiency roll-off. However, its implementation requires careful signal deconvolution (e.g., separating bleaching and Stark components) and often relies on model-based analysis for quantitative interpretation. These limitations highlight the need for integrated multi-modal and operando approaches to fully resolve charge dynamics and degradation pathways in practical QLED systems.

Aging mechanism & spatial heterogeneity

Electronic absorption spectrum. Electroabsorption (EA) spectroscopy is a valuable tool for probing internal electric fields in QLEDs and is therefore widely used to diagnose field-related efficiency losses, stability issues, and degradation pathways.¹⁰⁷ In a typical EA measurement, as schematically illustrated in Figure 9A, a small sinusoidal modulation voltage, $V_{ac} \sin(\omega t)$ (often 1 kHz), is superimposed on a DC bias V_{dc} , generating a periodically varying field across the device stack. The field induced a Stark response in functional layers (e.g., the emissive layer and HTL), leading to a field-dependent change in the absorption coefficient $\Delta\alpha(\lambda)$, which is detected optically by monitoring the normalized modulation of the probe intensity (e.g., $\Delta T/T$ or an equivalent returned signal). Using a monochromatic probe incident at $\sim 45^\circ$ and locked-in detection referenced to ω , the weak EA response can be selectively amplified; under injection-suppressed conditions the EA amplitude scales with E^2 , and the DC-AC cross term yields a component at the fundamental frequency, enabling quantitative tracking of the local field evolution within specific device layers.^{108–111}

Leveraging this capability, Luo et al.¹¹² directly determined the flat-band voltage (V_{FB}) of both the QD EML and HTL. As shown in Figure 9B, they showed that the QLED turn-on voltage ($V_{th} \approx 1.4$ – 1.5 V) closely matched the EML flat-band voltage ($V_{FB} \approx 1.41$ V), indicating that once the QD layer reached the flat-band condition, internal-field reversal strongly enhanced electron injection and triggered turn-on, supporting a classical injection-enhancement origin and excluding alternative explanations such as Auger up-conversion. Building on this capability, Zheng et al.¹¹³ exploited the linear dependence of the Stark signal on DC bias to quantify the built-in potential (V_{bi}) in QD photodiodes. They observed an increase of ~ 0.4 V in V_{bi} after aging ZnO nanoparticles for six days, providing direct evidence for a “positive aging” effect: improved energy-level alignment between ZnO and adjacent layers reduced the electron-injection barrier and facilitates electron transport, as depicted in Figure 9C.

By tracking the evolution of layer-specific flat-band and built-in potentials, EA spectroscopy has become a powerful operando tool to pinpoint the origin of operating-voltage rise (voltage loss) in QLEDs and linked it to underlying aging pathways. Sun et al.¹¹¹ quantified the impact of the hole-injection barrier on blue-QLED degradation by monitoring the built-in potential change in the QD emissive layer, ΔV_{bi} , during constant-current stress. After 3 h, ΔV_{bi} diverged markedly (0.03–1.07 V) across different electrode/HTL combinations: for a fixed electrode, deeper HTL HOMO levels led to larger ΔV_{bi} , whereas increasing the electrode work function strongly suppressed ΔV_{bi} , indicating that voltage loss is primarily governed by the HTL side. Moreover,

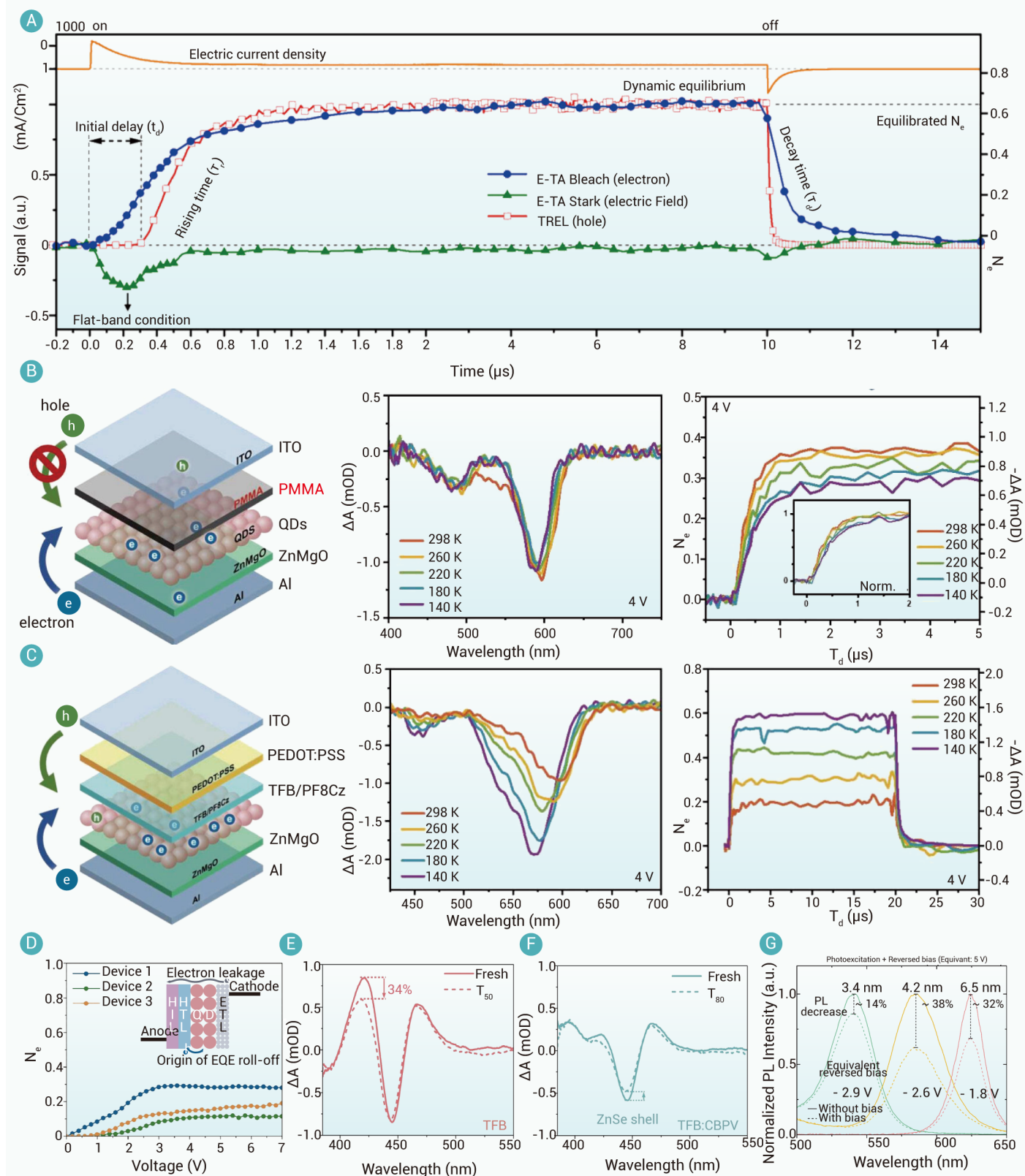


Figure 8. Electron and hole dynamics in QLEDs (A) Dynamics of electron and hole injection in a QLED, showing ETA bleach (electron), TREL kinetics (hole), and transient current (orange). The bleach signal is converted to electron density N_e , with equilibrium values indicating electron density in the QD layer. The Stark signal reflects the electric field evolution. Reproduced with permission.¹⁰⁰ Copyright 2024, American Chemical Society. (B) Structure of half-QLED with an insulating PMMA layer replacing the HTL to study electron injection (left), ETA spectra of the half at 4 V and various temperatures (middle), and electron population N_e from the bleach signal as a function of delay time T_d at 4 V at different temperatures (right).¹⁰³ Copyright 2025, American Chemical Society. (C) Structure of a conventional red QLED for hole injection analysis (left), ETA spectra of the half at 4 V and various temperatures (middle), and electron population N_e from the bleach signal as a function of delay time T_d at 4 V at different temperatures (right). Reproduced with permission.¹⁰³ Copyright 2025, American Chemical Society. (D) Average electron population $\langle N_e \rangle$ as a function of bias. Inset is that the possible origins for the efficiency roll-off in QD-LEDs. Reproduced with permission.¹⁰⁴ Copyright 2024, American Chemical Society. (E, F) ETA spectra of QLEDs using (E) TFB and (F) photo-crosslink TFB: CBPV as HTLs, respectively, before (solid) and after degradation (dashed) under constant driving at 5 V. Reproduced under the terms of the CC-BY license.¹⁰⁵ Copyright 2025, Li et al., published by Nature Communications. (G) PL spectra without (solid) and with (dashed) an equivalent external reversed bias (+5 V). reproduced with permission.¹⁰⁶ Copyright 2025, American Chemical Society.

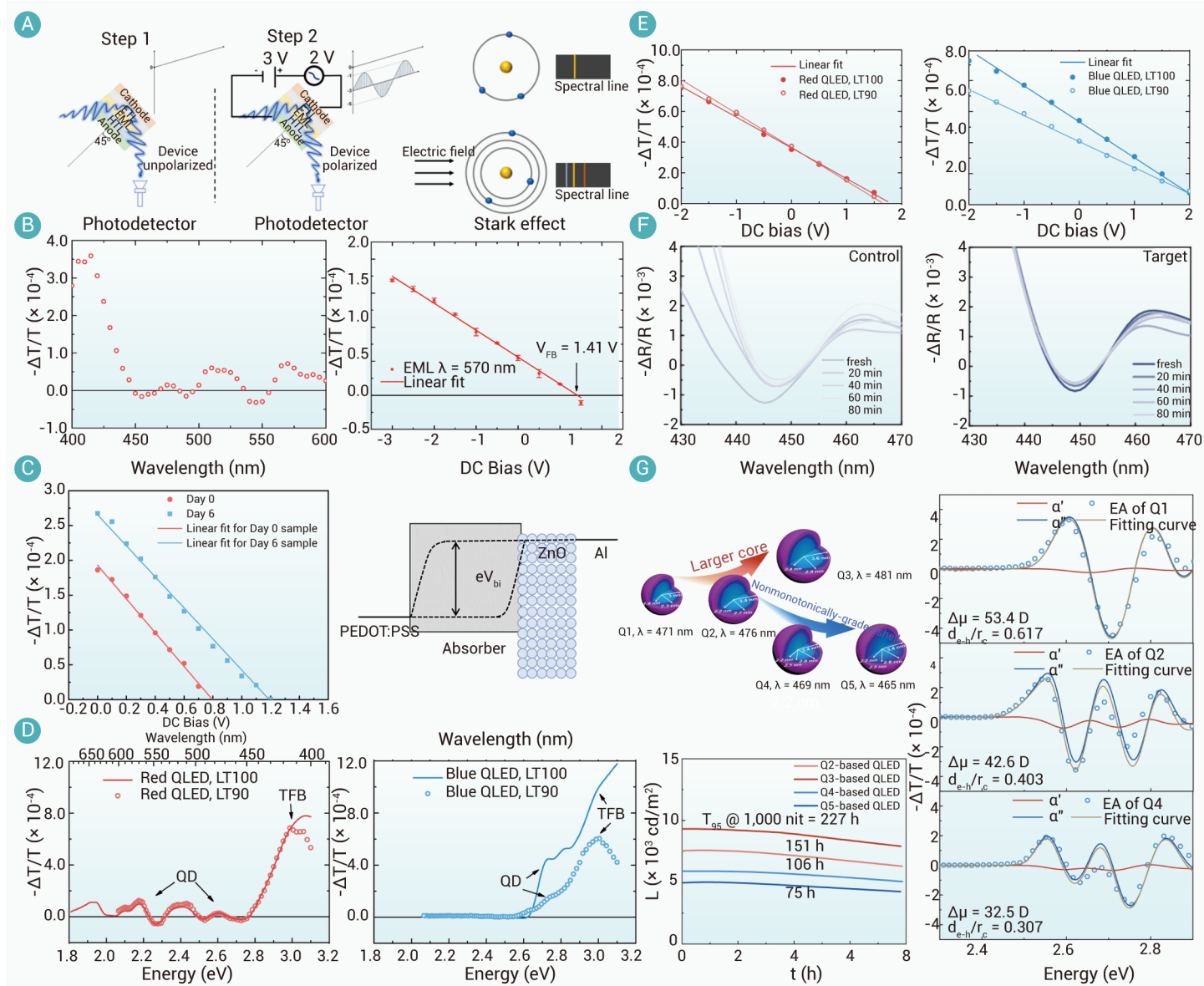


Figure 9. Electroabsorption (EA) analysis of internal electric field and device stability (A) Schematic of EA measurement of the device with/ without an applied electric field. Reproduced with permission. Copyright 2024, American Chemical Society.¹¹⁴ (B) EA spectrum of red QLEDs (left) and extraction of V_{FB} from the bias-dependent Stark response of red QDs (right). Reproduced with permission.¹¹² Copyright 2019, American Chemical Society. (C) Determination of V_{bi} using bias-dependent electro-absorption (left) and schematic diagram showing $eV_{bi} = \Phi_{PEDOT:PSS} - \Phi_{ZnO/Al}$ (right).¹¹³ Copyright 2020, American Chemical Society. (D) In-phase EA spectra of standard red and blue QLEDs (ITO/PEDOT:PSS/TFB/QD/ZnO/Al) measured at DC bias = -1 V and AC amplitude = 0.1 V, before and after lifetime tests; arrows indicate contributions from TFB and QDs.¹¹⁰ (E) Bias-dependent EA signals of TFB (probe wavelength fixed at 415 nm) in standard red and blue devices, reflecting the internal electric field across the HTL. Reproduced under the terms of the CC-BY license. Copyright 2019, Chen et al., published by Nature Communications.¹¹⁰ (F) EA spectra of control (left) and CI-treated QLEDs (right). Reproduced with permission.¹¹⁴ Copyright 2024, American Chemical Society. (G) Core/shell/shell structures for Q1-Q5; EA spectra and the extracted dipole moments of Q1, Q2, and Q4 measured on ITO/PEDOT:PSS/QDs/Ag (~15 nm); luminance decay of blue QLEDs based on Q2-Q5 operated at 50 mA/cm². Reproduced under the terms of the CC-BY license.³³ Copyright 2023, Chen et al., published by Nature Communications.

ΔV_{bi} closely tracked the operating-voltage rise ΔV_0 and showed the same dependence on the barrier configuration $\Delta V_1/\Delta V_2$, leading to a practical design rule – minimize the electrode/HTL interfacial barrier ΔV_1 to mitigate voltage increase and irreversible luminance loss, even if the HTL/QD offset ΔV_2 become large (up to ~0.8 eV). This behavior was further rationalized in terms of enhanced electrode-induced p-type doping in the HTL, providing a spectroscopic basis for stability optimization via HTL p-doping engineering. Complementarily, Chen et al.¹¹⁰ used EA to compare degradation pathways in red versus blue QLEDs. Although both devices showed a small population of oxidized species (TFB⁺) after aging, the HTL-related EA response (and associated voltage-loss evolution) changed only slightly, implying that HTL aging contributed mainly to the slow degradation of red devices but is not the dominant cause of the short lifetime in blue QLED (Figure 9D). Instead, EA revealed a pronounced out-of-phase (quadrature) charge-modulation signal at the blue QD/ZnO ETL interface, consistent with interfacial charge transfer

and accumulation as the primary driver of the increased voltage loss in blue devices (Figure 9E).

More broadly, EA spectroscopy is exceptionally sensitive to charge-related interfacial dynamics, enabling direct identification of carrier transfer and accumulation in QLED stacks. In the study of Chen et al.,¹¹⁰ mixed QD-ZnO stacks showed a pronounced quadrature charge-modulation signal for the blue QD-ZnO system, whereas the corresponding red QD-ZnO stack exhibited a negligible response. This contrast unambiguously indicated efficient electron transfer from blue QDs into ZnO followed by charge build-up in the ETL, which – together with bias-dependent EA and complementary C-V analysis – was linked to the voltage-loss increase predominantly originating from the ETL in blue devices. From a complementary perspective, Fei et al.¹¹⁴ highlighted the importance of suppressing charging within the QD emissive layer. Employing EA to monitor heavy-metal-free ZnSeTe blue QLEDs under constant current stress (100 mA/cm², 0–80 min) as shown in Figure 9F, they

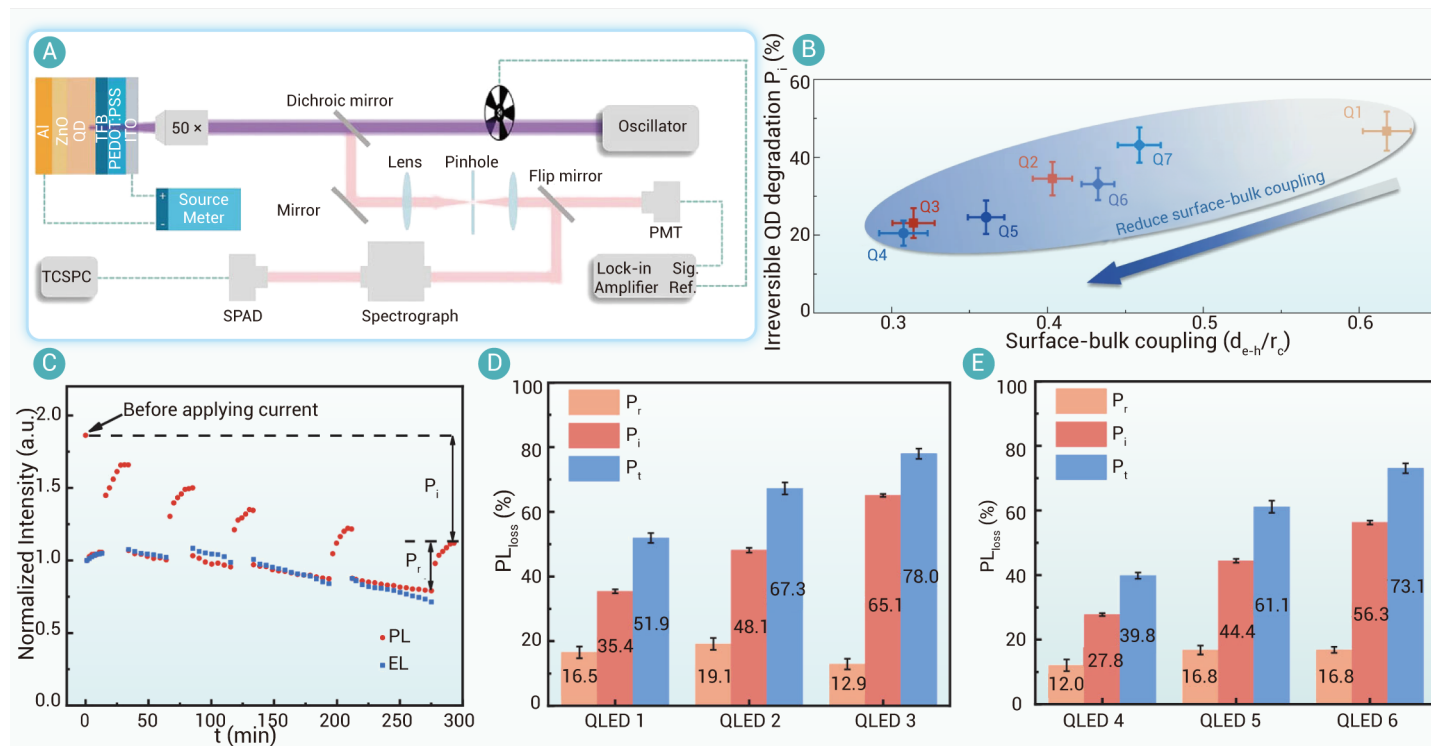


Figure 10. Surface-Bulk coupling and degradation mechanisms in QLEDs (A) schematic diagram of the EL and PL aging test. Reproduced under the terms of the CC-BY license.⁸⁸ Copyright 2024, Su et al., published by advanced Physics Research. (B) Correlation between surface-bulk coupling (d_{e-h}/r_c) and charge-induced irreversible degradation (P_i) for Q1–Q7 (error bars indicate data range). Reproduced under the terms of the CC-BY license.³³ Copyright 2023, Chen et al., published by Nature Communications. (C) Typical EL–PL test curve. Reverse bias pulses (rectangular wave: –10V, 0.5 Hz, 0.6 duty cycle) were applied intermittently to partially recover PL loss, with reversible and irreversible PL losses labeled. (D, E) Reversible, irreversible, and total PL loss for Devices 1–3 and Devices 4–6. Reproduced with permission.¹¹¹ Copyright 2024, American Chemical Society.

observed that the untreated control device showed a clear redshift and attenuation of the first excitonic feature, consistent with progressive charge accumulation/redistribution and field-induced spectral distortion. In contrast, devices treated with an inorganic X-type ligand ($\text{ZnCl}_2/\text{Cl}^-$) maintained an essentially invariant EA line shape, demonstrating that Cl^- ligation effectively mitigates operational charging. Consistently, the device lifetime improved markedly, with T_{50} increasing from 7.3 h to 52.7 h (~ 7 folds), underscoring ligand engineering as an effective route to enhance the stability of heavy-metal-free blue QLEDs.

Beyond its diagnostic role at device level, the EA spectral line shape encodes microscopic information on QD excitonic states, thereby enabling direct correlations between QD electronics structure and operational stability. Chen et al.³³ analyzed a conventional monotonic-gradient blue QD (Q1) and extracted an unusually large excitonic dipole moment ($\mu \approx 53.4$ D) together with a high normalized electron-hole separation ($d_{e-h}/r_c = 0.617$), indicative of strong surface-bulk coupling that amplified the influence of localized surface charges and accelerated degradation. In an optimized QD with a non-monotonic gradient shell (Q4), in an optimized QD with a non-monotonic gradient shell (Q4), d_{e-h}/r_c decreased to 0.307, demonstrating that shell engineering can effectively weaken this coupling. Consistently, time-dependence EA revealed pronounced, partially reversible spectral redshifts during operation 200 min for Q1 (33.6 meV at first exciton and > 100 meV near the band edge), consistent with charged-induced surface-related dark-state processes, whereas optimized QDs (Q2 – Q5) exhibited much smaller shifts (6.7–11.1 meV) and markedly improved stability, boosting T_{95} (@1000 cd/m^2) from 47h to 227 h (sky blue) and 75 h (deep-blue), as summarized in Figure 9G. EA spectroscopy provides a direct, layer-resolved optical handle on internal-field and charge-driven processes in operating QLEDs. By quantitatively extracting key metrics and distinguishing interfacial charge accumulation from intrinsic layer degradation, EA provides a direct link between microscopic field/charge dynamics and macroscopic voltage degradation and lifetime behavior. Consequently, EA-derived descriptors offer actionable guidelines for device optimization, including interfacial barrier engineering, transport-layer stabilization, ligand passivation, and QD structural design.

EA spectroscopy enables layer-resolved probing of internal electric fields and charge-related process in operating QLEDs, providing unique insight into field evolution, interfacial charge accumulation, and voltage-loss pathways. It allows quantitative extraction of key parameters such as flat-band voltage and build-in potential, and enables direct correlation between microscopic charge/field dynamics and macroscopic device degradation and stability. However, its interpretation often relies on model-based analysis and careful spectral deconvolution, and separating overlapping contributions from different layers can be challenging. These limitations highlight the need for complementary multi-modal and operando approaches to fully resolve charge-field interactions in practical QLED systems.

Nanosecond transient absorption spectroscopy. When combined with time-resolved photoluminescence (TRPL), nanosecond transient absorption (ns-TA) spectroscopy could provide a particularly powerful mechanism-specific probe for interfacial exciton quenching in QLEDs because it could discriminate between electron-transfer and hole-transfer pathways. Both electron and hole transfer shorten the TRPL lifetime by introducing additional nonradiative decay channels. In ns-TA, however, electron transfer accelerated the recovery of the band-edge bleach, whereas hole transfer could leave the bleach minimally changed. Thus, the fast PL but low TA signature is a practical indicator of hole-transfer-dominated quenching.^{115,116} This approach has been applied to rationalize and mitigate QD/ETL interfacial losses. Zhang et al.¹¹⁶ showed that interfacial quenching depended strongly on the oxide ETL: a near-monolayer CdSe–CdZnS QD film in direct contact with ZnO exhibited pronounced quenching (PLQY $\sim 47\%$), whereas replacing ZnO with $\text{Zn}_{0.9}\text{Mg}_{0.1}\text{O}$ suppressed quenching (PLQY $\sim 68\%$) and prolonged the transient decay, indicating that Mg doping “softened” the QD/ETL interaction and enabled insulating-layer-free architectures. This strategy translated to green devices, boosting peak EQE from $\sim 11.1\%$ to 18.1%.

Employing the combined ns-TA/TRPL criterion, Chen et al.¹¹⁷ further identified the dominant loss channel at QD/ $\text{Zn}_{0.9}\text{Mg}_{0.1}\text{O}$ interface as hole transfer (slower TA but faster TRPL). They then introduced strongly quantum-confined “optical ZnO” (O–ZnO), which restored the TA and TRPL decays of the QDs to near-identity with those of pristine QD films – evidence that

Table 2. Advanced characterization methods for QLED device physics.

Method	Scale	Observable	Key metrics	What it reveals	Why it matters	Limitation/Challenge
Single-dot PL	Single QD	PL trace; $g^2(\tau)$	on/off; Auger; QY	Charging-induced blinking; Auger loss	Intrinsic stability limits; Anti-blinking design for high EQE	Limited representation of ensemble/device behaviors
Single-dot EL	Single QD (electrical)	EL; $g^2(0)$	$g^2(0)$; k_e/k_{nr} ; T_x	Injection asymmetry; Charged pathways	Exciton-formation limits; stability under bias	Specialized devices structures; low signal throughout
ETA	Device	ΔA (bleach + stark)	$\langle N_e \rangle$; delay; QCSE	injection/ accumulation vs leakage vs QCSE	Efficiency roll-off mechanisms; confinement/ injection design	Challenging signal separation
TREL/TRC	Device	TREL and TRC	t_d ; τ_r ; τ_d ; RC	Injection/ storage/ recombination kinetics	Charge imbalance diagnosis; transport process optimization	RC delay effects and coupled charge-carrier processes
Electro absorption	Device	Stark-modulation	Internal field; V_{FB}/V_{bi} ; μ_{exc}	Layer field evolution; interfacial charging	voltage loss pathways; degradation mechanisms	Model-dependent analysis and calibration requirements
ns-TA	Film	Bleach recovery & PL decay	τ_{TA} (with τ_{PL})	ET- vs HT- dominated quenching	Interface loss mapping; targeted passivation strategies	Optical excitation and limited relevance to electrical operation
Single-carrier device	Device	J-V and C-V (EOD/HOD)	J injection capacitance feature	Injection asymmetry; charge-balance limitation	Charge-balance engineering for EQE/ luminance/ lifetime	Simplified structure and limited representation of real operation
In-situ EL/PL	Device	EL decay and modulated PL	P_r ; P_i	reversible charging vs irreversible damage	Failure-mode identification; lifetime analysis	Complex experimental setup and challenging signal separation
Spatial resolved mapping	Pixel	Time-gated EL maps	Local intensity	Nonuniform injection; hotspots	Spatial heterogeneity-degradation correction; uniformity control	Faced trade-offs between spatial/ temporal resolution trade-off and advanced instrumentation

the hole-transfer pathway was effectively blocked – raising the PLQY of a QD monolayer on O-ZnO to ~ 74%. Luo et al.¹¹⁸ extended this framework via comparing QD/O-ZnO with amine-functionalized O-ZnO (O-ZnO-NH₂). QD/O-ZnO again exhibited the “fast PL/slow TA” signature, whereas O-ZnO-NH₂ produced synchronized TA and TRPL decays close to pristine QDs, confirming that amine functionalization suppressed hole-transfer quenching. Correspondingly, PLQY improved from ~ 39% to ~ 75% (green) and from ~ 31% to ~ 70% (blue), with average lifetimes extended, 7.5 to 16.4 ns and 8.7 to 13.7 ns, respectively, enabling peak EQEs of 22.4% (green) and 14.3% (blue) and maintaining essentially unchanged performance after 270 days of storage. ns-TA spectroscopy combining with TRPL has become a widely adopted approach for resolving the microscopic origins of interfacial exciton quenching in QLEDs. By distinguishing whether nonradiative loss is dominated by electron transfer or hole transfer, this paired analysis enables rational, mechanism – guided interface engineering. Such targeted strategies suppress interfacial nonradiative pathways and translate directly into improved device efficiency and operational stability.

ns-TA coupled with TRPL enables mechanism-specific probing of interfacial exciton quenching in QLEDs by distinguishing electron- and hole- transfer pathways. It provides unique insight into the origin of nonradiative losses through correlated analysis of bleach recovery and PL decay dynamics, allowing direct identification of dominant interfacial quenching mechanisms. However, its interpretation relies on comparative analysis of coupled spectroscopic signals and is typically limited to model systems rather than fully operating devices. These limitations highlight the need for complementary operando and device techniques to fully capture interfacial charge-transfer processes in practical QLEDs.

In-situ electroluminescence and photoluminescence. Operando in situ EL/PL measurements provide a direct way to track EL and PL simultaneously in operating QLED, thereby linking emission decay to interfacial and material degradation.^{107,119} The method is built on lock-in detection: EL generated under constant-current driving appears as a DC background, while a chopped monochromatic excitation produced a frequency-modulated PL component; referencing the lock-in to the chopper isolated PL, and the

remaining DC signal corresponded to EL (Figure 10A). By monitoring the evolution of PL under electrical bias and during reverse-bias recovery, the total PL loss (P_i) can be quantitatively separated into reversible and irreversible components. When further combined with reverse-bias recovery, the PL loss (P_i) could be divided into a reversible part (P_r , transient charging/ tripping) and an irreversible part (P_i , permanent QD/interface degradation), enabling mechanistic failure diagnosis.^{33,120-122} Using this strategy, Chen et al.³³ compared the conventional gradient-shell blue QD (Q1) with structurally optimized QDs (Q2 – Q5), as summarized in Figure 10B. They found that P_i was dominating in life extension: structural QDs: with structural optimization, P_i could be reduced from conventional QDs 46.7% to optimized QDs ~ 20 – 24%, and the device with the smallest P_i could achieve the longest lifetime ($T_{95} = 227$ h @ 1000 cd/m²). As shown in Figures 10C-E, Sun et al.¹¹¹ further showed that different electrode/HTL combinations exhibited similar P_i values (~ 10-20%), whereas P_r varied widely (27.8% – 65.1%). P_i was primarily governed by the electrode/HTL hole-injection barrier (ΔV_1): a large ΔV_1 led to more severe irreversible loss. Increasing the electrode work function to reduce ΔV_1 could suppress P_i , even if it resulted in a larger nominal HTL/QD offset ΔV_2 . Therefore, materials selection should prioritize electrode/HTL energy-level matching (minimizing ΔV_1) to mitigate fixed-charge accumulation and reduce irreversible degradation. Shen et al.¹²⁰ extend this in-situ EL/PL methodology to NIR-QLEDs by comparing devices with a conventional LiF interlayer to those employing ETL:Li composite layer. Over 150 min of operation, the LiF-based devices exhibited an ~ 12 % decrease in EL intensity, whereas the ETL:Li devices showed essentially no EL degradation; the PL intensity of them remained unchanged. These results indicated that the performance decay was dominated by an additional cathode/ETL interfacial quenching pathway, rather than the intrinsic instability of QD EML. By eliminating this interface and introducing the Li-doped ETL, the V_{on} voltage was reduced the EQE increased by ~ 3-fold and the operational lifetime improved markedly from $T_{80} = 5.3$ h to 32 h @ 1 mA/cm², underscoring the decisive role of interface engineering in improving stability. This distinction between P_r and P_i is critical for identifying failure modes in QLEDs, P_i -dominated loss indicated transient processes such as charge accumulation and exciton quench-

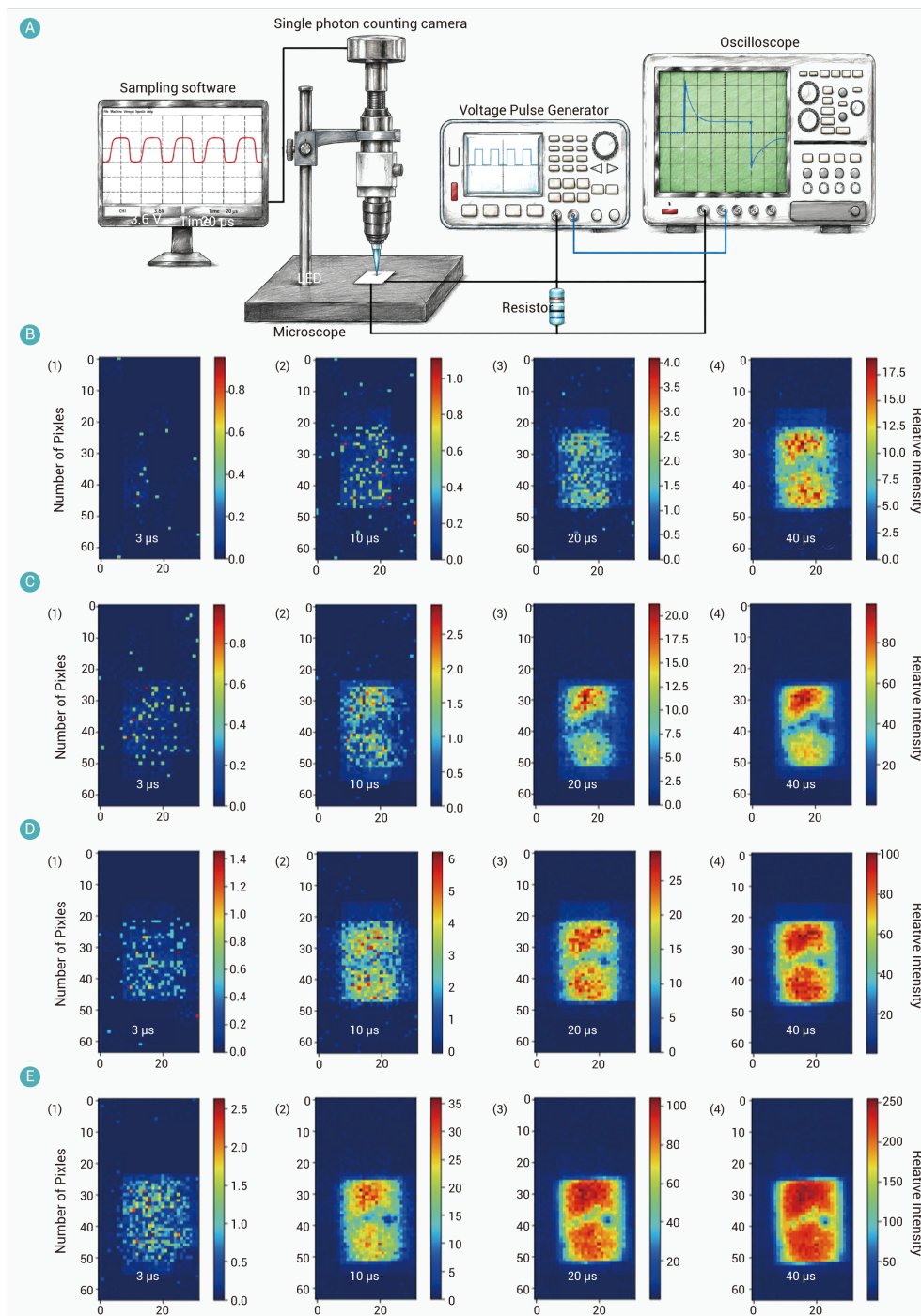


Figure 11. TSR-EL characterization of QLEDs (A) Schematic of the TSR-EL setup consisting of a voltage-pulse generator, oscilloscope, microscope, and SPC camera.¹²³ Time-resolved pseudocolor EL maps recorded at pulse widths of 2, 10, 20, and 40 μs under different driving voltages and temperatures: (B) 3.0 V, 293 K; (C) 3.0 V, 363 K; (D) 3.4 V, 293 K; (E) 3.4 V, 363 K. Reproduced with permission.¹²³ Copyright 2024, American Chemical Society.

signal separation, and its quantitative interpretation can be affected by device conditions. These limitations highlight the need for complementary operando approaches to fully resolve degradation mechanisms.

Spatially resolved microscopy. Spatially resolved imaging is a central component of time and spatially resolved electroluminescence (TSR-EL), which integrates a single-photon camera (SPC) with time-gated acquisition to reconstruct where and when light is emitted during operation, achieving a temporal resolution of ~ 10 ns and demonstrating a spatial resolution of ~ 126 μm on GaN-based micro-LEDs, underscoring the method's broad applicability, as shown in Figure 11A. Benefiting from the extremely high sensitivity of SPC, spatially resolved imaging could detect very weak emission (~ 20 cd/m²) near turn-on voltage that is often inaccessible to convention APD/PMT measurements, providing spatial pseudo-color maps recorded under different drive condition (e.g., 3.0 vs 3.4 V and 293 vs 363 K) reveal a clear trend toward improved EL uniformity with higher voltage, elevated temperature, and longer injection time, as shown in the Figures 11B-E – evidenced by reduced pixel-to-pixel intensity variation – consistent with increased charged accumulation and supported by TRC analysis.¹²³

Moreover, TREL-based imaging can extract pixel-resolved parameters such as brightness, rising time, and decay time across degradation stages, revealing that non-uniform electron injection initially caused brightness inhomogeneity; the distribution became more uniform during positive degradation but remains heterogeneous during negative degradation.⁹⁶ TSR-EL provides a uniquely sensitive, nanosecond-resolved view of spatiotemporal emission dynamics. By linking EL maps with TRC/TREL

ing, whereas P_i -dominated loss reflected intrinsic material or interfacial degradation. In-situ EL/PL measurements have become a key bridge in QLED research, linking materials design, device engineering, and ultimate performance. This approach provided a unique capability to decouple photoluminescence loss into reversible (P_r) and irreversible (P_i) components, thereby distinguishing transient charge-induced quenching from permanent material and interfacial degradation. Such separation offered direct insight into the dominant failure modes of QLEDs and established clear guidance for device optimization. Importantly, suppressing P_i was critical for extending device lifetime, enabling targeted optimization both materials and architectures.

In-situ EL/PL measurements enable simultaneous tracking of EL and PL in operating QLEDs, allowing PL loss to be decoupled into reversible (P_r) and irreversible (P_i) components. This provides direct insight into transient charge-induced quenching versus permanent material and interfacial degradation, enabling clear identification of failure modes and their impact on device lifetime. However, the technique requires complex lock-in detection and careful

metrics, it shows that charge accumulation and injection nonuniformity drive pixel-to-pixel brightness variations and their changes during degradation.

Spatially resolved TSR-EL imaging enables visualization of spatiotemporal emission dynamics in operating QLEDs, revealing local charge injection and transient behavior with high sensitivity. However, its spatial-temporal resolution is limited by instrumental trade-offs, and quantitative analysis can be complex. These limitations highlight the need for advanced and complementary techniques to fully resolve local degradation processes.

Advanced characterization techniques in this section provide complementary insights into QLED aging from multiscale perspective. EA probes internal electric fields and interfacial charge accumulation, while ns-TA coupled with TRPL resolves interfacial carrier transfer pathways at the nanoscale. In-situ EL/PL further distinguishes the reversible and irreversible degradation, and the local heterogeneity in carrier injection and emission could be revealed by spatial resolved mapping. These approaches are intrinsically interconnected, linking microscopic processes such as charge accumulation, interfacial

transfer, and exciton quenching to macroscopic device degradation and nonuniformity. Integration of insights across different techniques and scales collectively constructs a coherent, mechanism-driven framework to understand QLED aging and guide device optimization.

Taken together, the advanced characterization methods summarized here (Table 2) establish a hierarchical operando diagnostic framework for QLEDs, spanning intrinsic single-emitter stability limits, devices-scale charge/field dynamics, and spatially heterogeneous aging. By mapping observables to quantitative metrics, these tools enable mechanistic discrimination among charge imbalance, carrier leakage, QCSE-driven field effects, and interfacial/bulk degradation-loss channels that are often indistinguishable using conventional steady-state electrical and optical measurements. Among them, ns-TA provides a representative example of how operando spectroscopy can quantitatively separate carrier injection/accumulation from internal field responses, thereby linking microscopic dynamics to actionable design rules in confinement and injection control.

CONCLUSION AND OUTLOOK

The highly developed methods of characterization that were employed in investigating QLEDs have greatly improved our understanding of the fundamental processes involved in the operation of QLEDs, as well as their efficiency and degradation modes. These techniques allow detailed observations of charge injection, exciton dynamics, and interface stability, providing a more comprehensive understanding of the factors that determine QLED operation. A combination of a variety of methods of characterization would allow the investigators to further narrow the range of factors responsible for efficiency roll-off, long-term instability, and other challenges that QLEDs currently encounter. These methods are invaluable in the investigation of the spatial inhomogeneity in emission, the understanding of the influences of the charge dynamics on the degradation, and the establishment of correlations between materials properties and the characteristics of the devices. The coincidence of both high spatial and temporal resolution is such that a comprehensive study of QLED behavior under a wide range of operational conditions is possible to reveal important processes that have been inaccessible by direct observation in the past.

Despite the remarkable progress in QLED performance, several key challenges still remain, particularly the coupled processes of charge transport, exciton recombination, and interfacial effects that occur on ultrafast timescales and at localized regions. Therefore, future characterization should focus on techniques with high spatiotemporal resolution, enabling direct correction between transient dynamics and local heterogeneity in operating devices. Particularly for blue QLEDs, which suffer from significantly inferior stability, advanced methods with high spatiotemporal resolution are urgently needed to probe high-energy carrier dynamics and local degradation. Techniques combining electrical excitation with ultrafast and spatially resolved detection will be critical, while machine-learning-assisted analysis will be essential for extracting physically meaningful descriptors and decoupling complex processes. This perspective highlights the direction for next-generation operando characterization platforms.

Future developments will accrue through operando diagnostics driven by workflows: (i) employ single-carrier diagnostic devices to detect injection asymmetry and the charge-balance bottleneck; (ii) apply TREL/TRC to quantify injection decay, charge storage, and recombination kinetics under pulsed operation; (iii) deploy ETA to separate carrier accumulation/leakage from QCSE to efficiency roll-off; and (iv) use EA to track layer-resolved field evolution and voltage-loss pathways during aging. Simultaneously, localization of hot integrating spatially-resolved time-gated mapping. Furthermore, it is expected that machine-learning-aided analysis will be increasingly useful to extract physically meaningful and robust descriptors of many multimodal operando datasets (e.g., TREL/TRC waveforms, ΔA spectra, EA-derived field profiles, and spatial maps), and hence automated classification of mechanisms, early-failure detection, and expedited discovery of the exciting loss channel across materials stacks. Coupled with high-throughput synthesis/processing and constrained optimization guided by physically meaningful measures, this approach will enable mechanism-inspired methods if engineering confinement strategies (e.g., shell/barrier design), interfacial passivation/ligands, and transport-layer energetics, ultimately bridging

laboratory performance to manufacturable lifetime scales and uniformity targets.

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AUTHOR CONTRIBUTIONS

Y. Zhao and Y. Liu collected the data and conducted the literature review for this work. Y. Zhao drafted the manuscript. B. Wu and S. Chang conceptualized, reviewed and edited the manuscript. The other authors revised the text to ensure the accuracy and completeness of the study. All authors contributed to the manuscript and approved the final version.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

No new data were created or analyzed in this study.